

THE SWELLING PRESSURES DEVELOPED BY SOME NEW ZEALAND COALS  
DURING CONSTANT VOLUME CARBONISATION

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by

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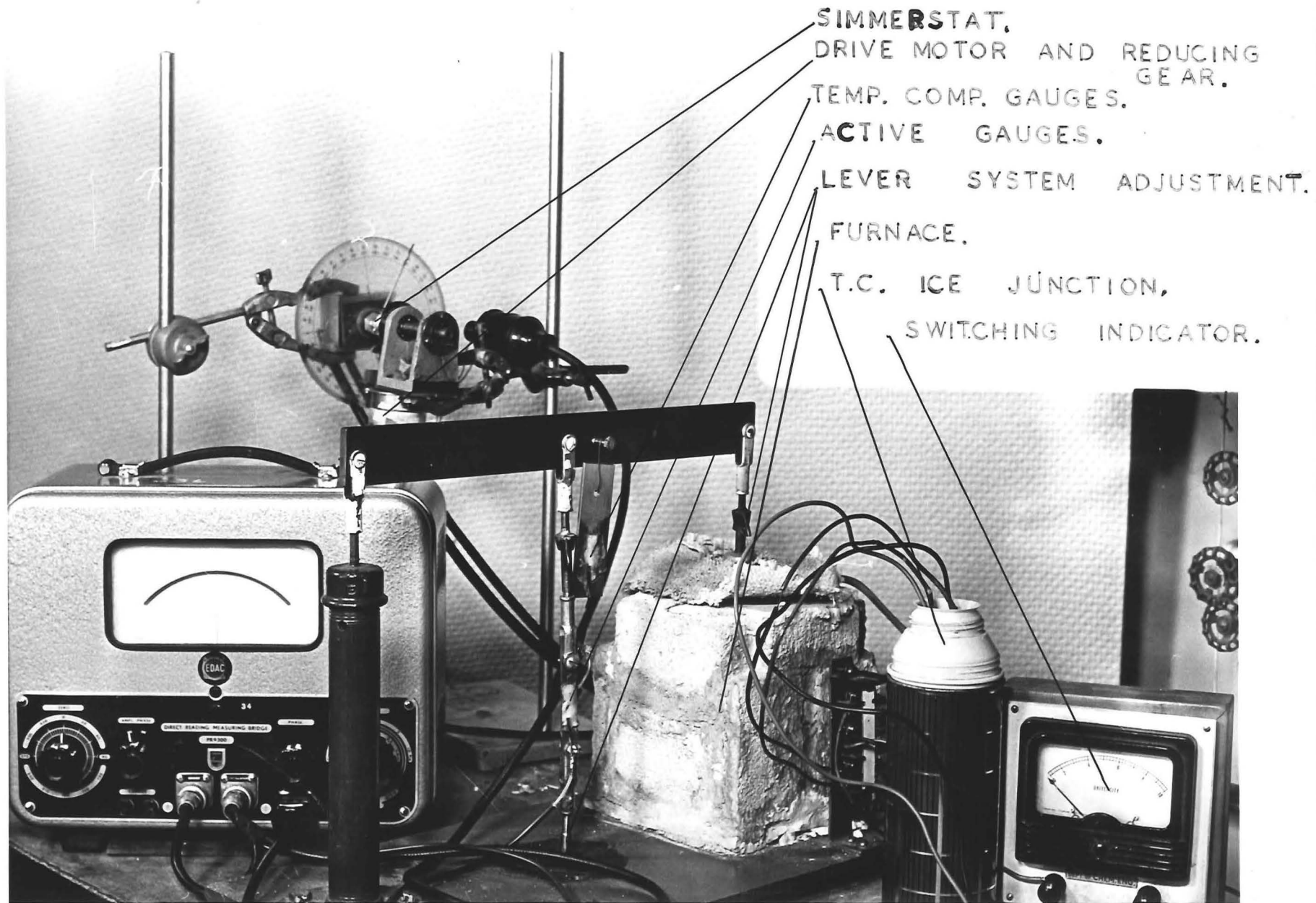


PLATE I

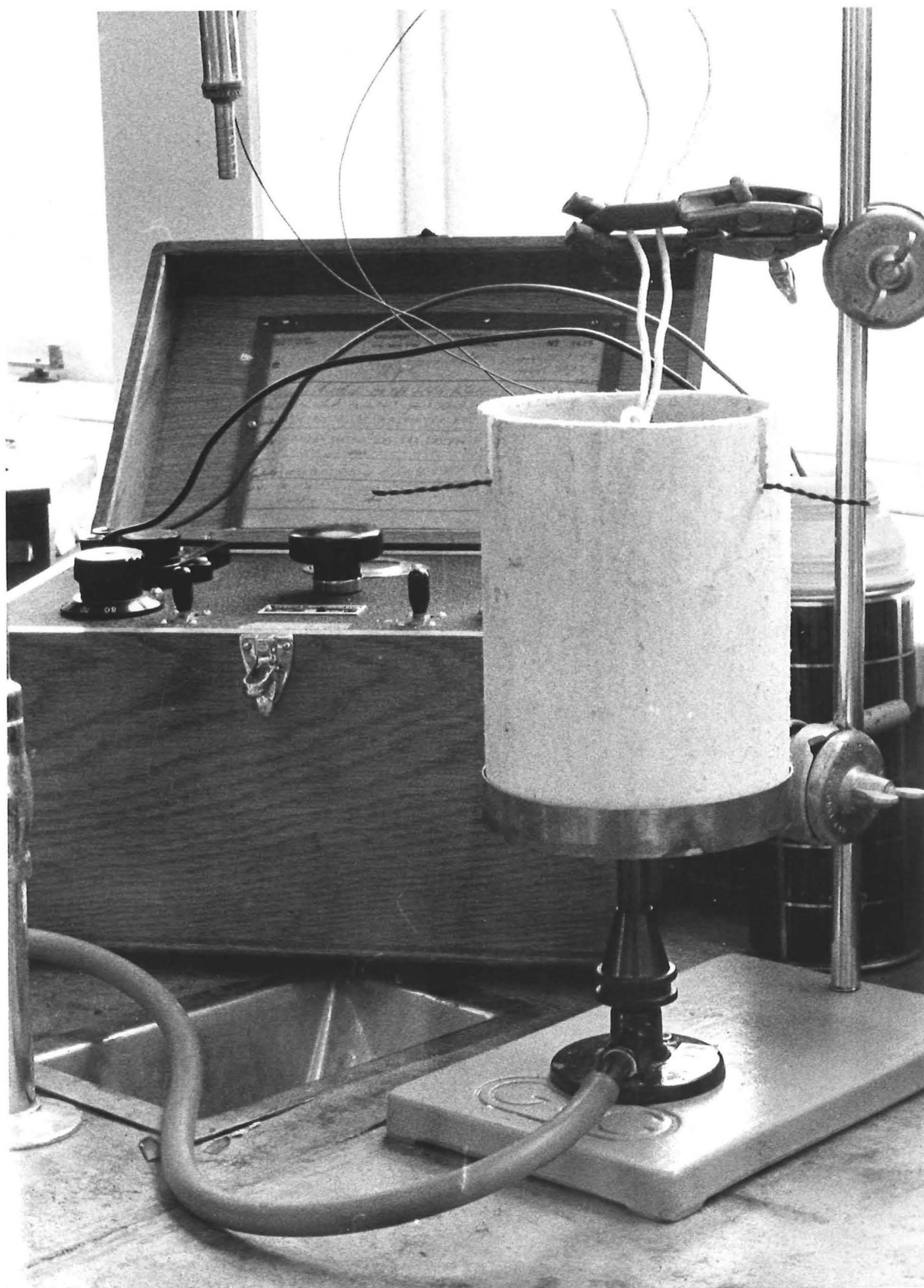


PLATE 2

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## SUMMARY

A dilatometer type apparatus has been used to measure the swelling pressure developed in a small coal sample during carbonisation at constant volume. A standard test procedure is outlined and the apparatus specified. Two types of results are given. In the first no corrections are made for effects not associated with the coal. These results, characteristic of the apparatus and standard test conditions, can be used as a quantitative measure of the swelling properties of coals, and are reproducible to  $\pm 5\%$ . In the second set of results elimination of such effects is attempted. The values so obtained are for the pressure at a point in a coal change throughout carbonisation.

These results show that swelling pressure occurs at lower temperatures than the associated property of volume increase, and is not due to volatile evolution through the plastic coal as the latter property is. The temperature(s) at which the maximum swelling pressure occurs is rank dependent and has a range 200-350°C in the rank range investigated. The magnitude of this maximum swelling pressure also increases with rank.

It is also shown that considerable shrinkage occurs in the coal sample after the maximum swelling pressure is developed.

The results are correlated with the BSS 1016 Crucible Swelling Test Scale and experimental data is extrapolated in an attempt to obtain equivalent swelling number values for the arbitrary 9<sup>+</sup>, 9<sup>++</sup>, 9<sup>+++</sup> designations at present used for high-swelling New Zealand coals. Values of 11½, 13½, 16½ respectively are predicted. Prediction of equivalent swelling numbers is also made using a percentage swelling - ultimate analysis formula. Similar values are obtained.

The correlations show swelling properties to have a second order dependence on swelling number. The results before correction for extraneous effects show an exponential dependence on swelling number.

### ACKNOWLEDGEMENTS

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I

INTRODUCTION

While a considerable amount of work has been done and continues to be done on the investigation of New Zealand coals, the greater proportion of this work is related to technological problems of utilization. Any information concerning fundamental coal properties has been incidental to solving the particular utilization problem encountered. Such was the case in the comprehensive survey of the dominion's coal resources undertaken by the Geological Survey and the Mines Department for which the Dominion Laboratory carried out sample analysis.

Suggate (1) in his D.S.I.R. Bulletin considers the results of this analytical work objectively and quite extensively in relating the properties of the coals found in New Zealand to their geological setting and history. He shows the unique nature of our coals with respect to overseas coals and shows that certain differences must be recognised when results of overseas research are used in considering New Zealand coals. For this reason a systematic investigation of fundamental New Zealand coal properties is desirable and offers much scope for research.

The present work was initiated to investigate a problem in utilization, namely the problem of excessive swelling and pressure development during carbonisation. The unique position of New Zealand coals in any consideration of the world coal resources is nowhere more evident than in the related properties of swelling (volume expansion during heating at constant pressure) and pressure development (during heating at constant volume). Certain New Zealand coals show swelling properties greater than those encountered in coals from any other world coalfield.

An arbitrary description of these swelling properties has been attempted



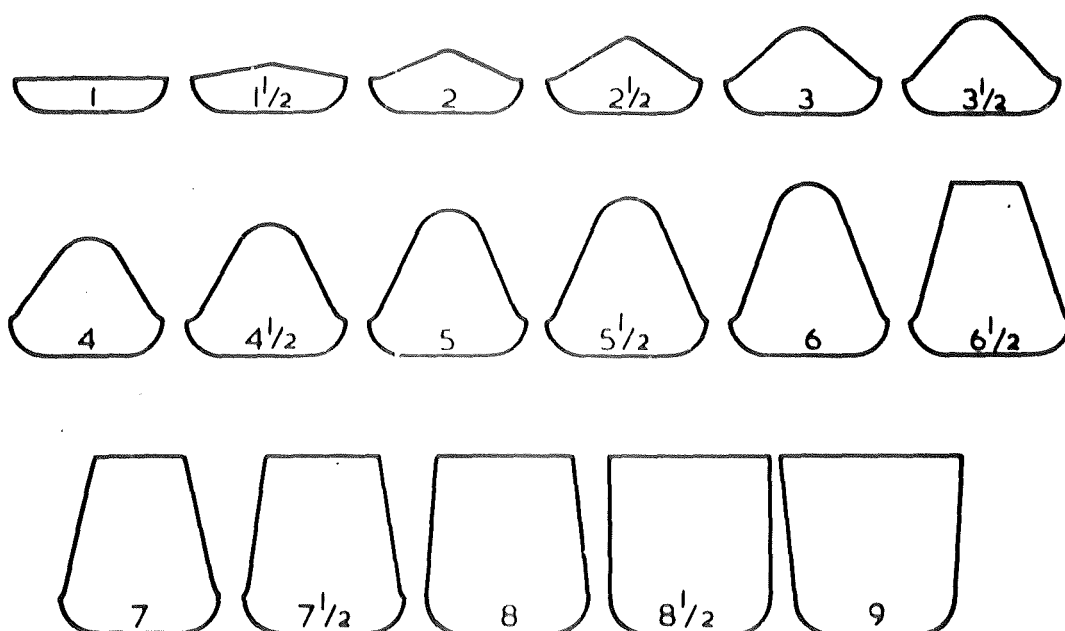


FIG. 1. Crucible Swelling Test — Standard coke button Profiles. Swelling numbers 0-9.

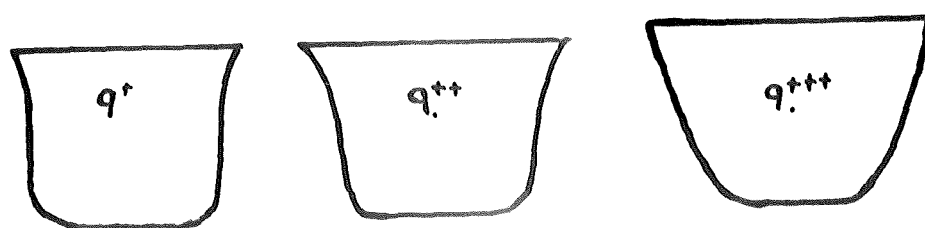


FIG. 2. Arbitrary Button Profiles — extension to range of FIG. 1.

in terms of the well established BSS 1016 Crucible Swelling Number test which defines swelling properties by means of the profiles of the coke buttons formed by carbonisation under the standard conditions described in BSS 1016 (2). The profiles designated 1-9 (fig. 1) represent a progressive percent volume increase in the coal sample carbonised in the test crucible. The arbitrary profile descriptions  $9^+$ ,  $9^{++}$ ,  $9^{+++}$  for coals with swelling properties greater than those represented by 9 on the standard scale are shown in fig. 2 and are due to the Coal Research Section of the Dominion Laboratory of the N.Z. D.S.I.R. (3). They represent evenly spaced increases in volume between the coke button 9 and a completely filled crucible  $9^{+++}$ .

For buttons  $> 6\frac{1}{2}$  it can be seen that free swelling is to some extent restricted, and that this restriction becomes more critical in the  $> 9$  region is shown by the lip on buttons  $9^+$  and  $9^{++}$ .

Therefore while giving a reasonably reproducible qualitative measure of the swelling properties of a coal the crucible swelling test cannot be considered to give a quantitative measure of swelling for coals of button numbers greater than  $6\frac{1}{2}$ . Moreover, for the particular problem under consideration the pressure developed by a coal charge during carbonisation was the factor of interest and the relationship of pressure developed to crucible swelling number for any coal is not known. Thus the crucible swelling test was unsatisfactory on two counts: it did not bear any known relationship to pressure properties so no absolute pressure values could be deduced from crucible swelling tests, and it gave no adequate quantitative indication of relative swelling or pressure developing properties between coals of different crucible swelling test button numbers.

As both absolute and relative pressure developments during carbonis-

ation were required for New Zealand high-swelling coals, investigation of a satisfactory test was undertaken. The choice of apparatus type, its construction and initial operation are considered in an earlier report (4).

Since that report some apparatus modifications suggested have been made and the first phase of the correlation of test results with the crucible swelling test scale has been attempted. It has been found that the results from the modified apparatus have been reasonably reproducible for the experimental conditions and are of interest not only with respect to the specific utilization problem which initiated the work but also for the information they give of the pressure and swelling properties in general for the limited range of coals considered. In many ways the results presented in this thesis are inadequate in number and not sufficiently reproducible for definite conclusions on the general behaviour of New Zealand coals to be made, but they contain indications of trends in pressure and swelling properties which are in some cases surprising and point the way to the most interesting areas for further more detailed investigation of these fundamental coal properties. Further work is being carried out in this field, initially on the utilization problem and pretreatment before carbonization (4). While this work will give further general information more basic research into fundamental coal properties along the lines suggested in section VII is desirable if any real understanding of the swelling pressure and associated physical properties of our coals is to result.

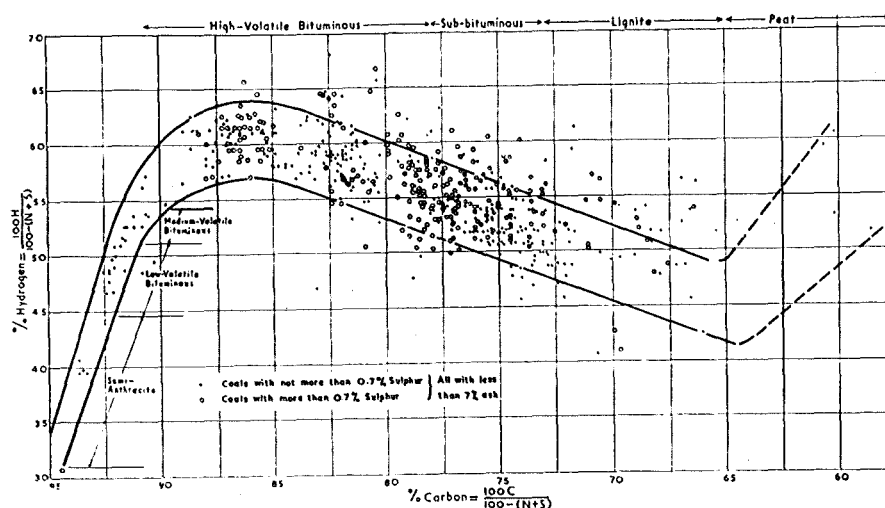


Fig. 3. The New Zealand coal band—axes of hydrogen and carbon.

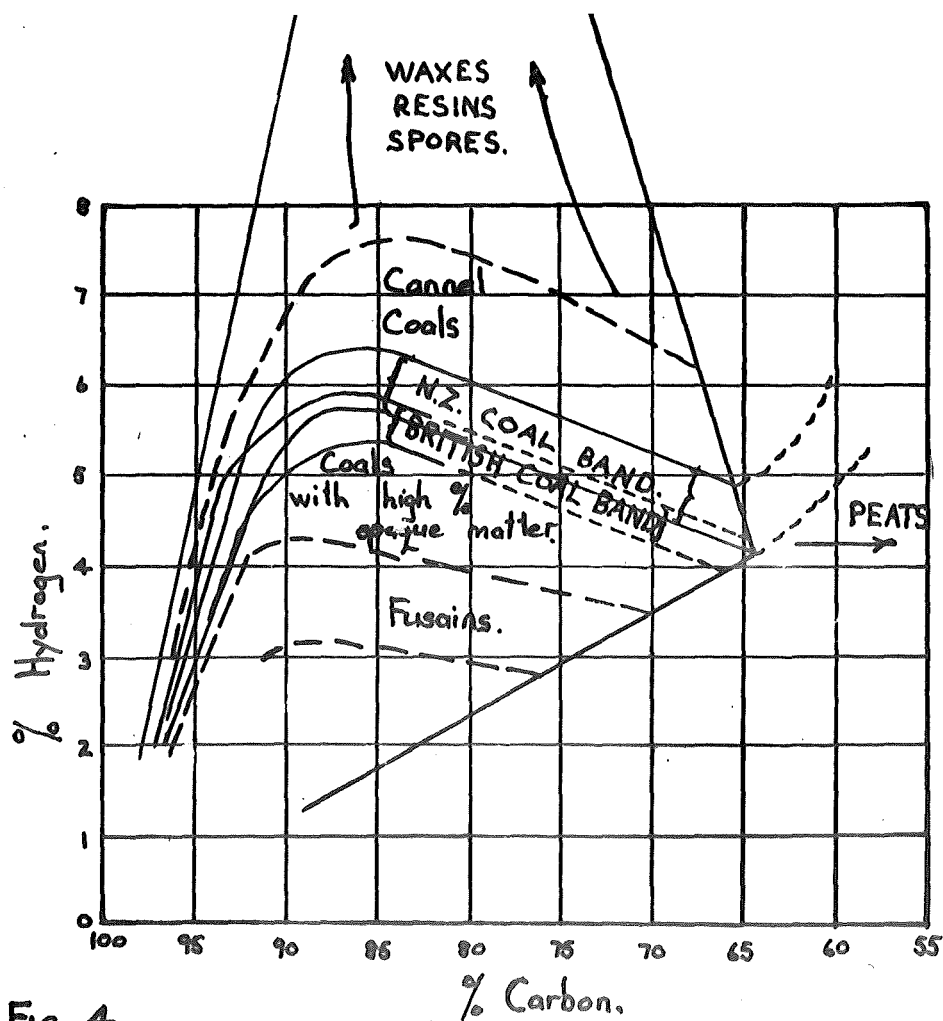
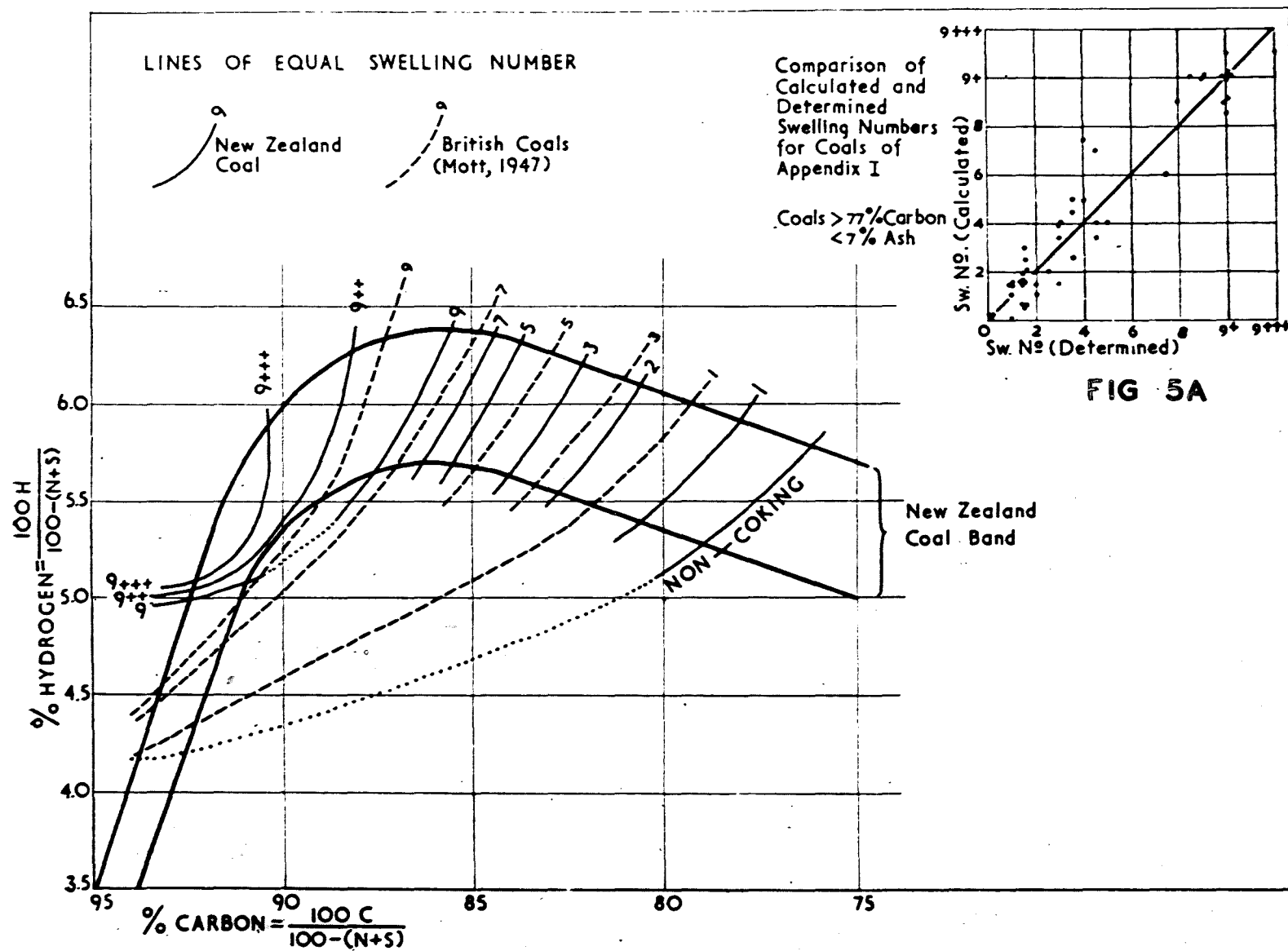


Fig. 4.  
(Fig. 34. SUGGATE).



## II

LITERATURE

The earlier report on this work (4) includes a comprehensive survey of work in the field of coal swelling and pressure developing property determination. Results are compared with those given by workers there referred to where comparison is legitimate. As previously mentioned, New Zealand coals are in many ways unique making comparison difficult. Much of the work considered in ref. 4 is to do with British coals which are carboniferous and orthohydrous. New Zealand coals are, in comparison, younger geologically (Cretaceous and Tertiary) and of distinctive type.

Suggate (1) gives the New Zealand coal band plotted on axes of percent hydrogen and percent carbon as shown in fig. 3 (fig. 9 in Suggate's work). The displacement of the British coal band with respect to the New Zealand coal band on these axes is shown in fig. 4, which is taken from Suggate's fig 34. As much of the comparable literature is based on work on British coals, this distinction between bands is relevant and important when comparison is being made.

Mott (5) presents formulae relating both percentage swelling and crucible swelling test numbers of British coals to the coal band. Fig. 5 (Suggate, fig. 24) shows lines of equal swelling for New Zealand coals plotted on the New Zealand coal band and Mott's lines of equal swelling for British coals are also shown. Suggate also shows the correlation of calculated and determined crucible swelling numbers for the coals whose analyses he considered, fig. 5(a).

It seems from Mott's relation for British coals as plotted that New Zealand coals not only develop much greater swelling properties than British coals but also that swelling properties develop at a lower carbon percentage

and the rate of development with increase in carbon percentage differs between the two coal bands. The double intercept of the lines of equal swelling number with the coal band is also of interest, especially in the Paparoa mine which yields coals in the region of the band where rapid decrease of swelling number with increasing carbon percentage occurs. Suggate shows that this could be due in part to increasing moisture content with decreasing volatile content below 23% and ease of acceptance of moisture of these coals. These factors possibly reverse the trend of resistance to weathering with rank increase in coals >23% volatile matter.

Spooner (6) gives a revised formula relating percentage swelling to ultimate analysis

$$\sqrt{\% \text{ swelling}} = \frac{1000 (H - 4.2)}{(O + 1)^2}$$

derived by comparing percentage swelling with crucible swelling number, again for British coals.

Suggate (1) shows that if Spooner's formula is applied to the average-type New Zealand coals the figures for percentage swelling at given crucible swelling numbers correspond fairly closely with those given by Spooner for British coals (see table 1). If the values given by Spooner's formula for New Zealand coals of crucible swelling numbers >9 are a proper indication of relative swelling properties, then the inadequacy of the crucible swelling test for quantitative assessment of swelling properties is immediately apparent.

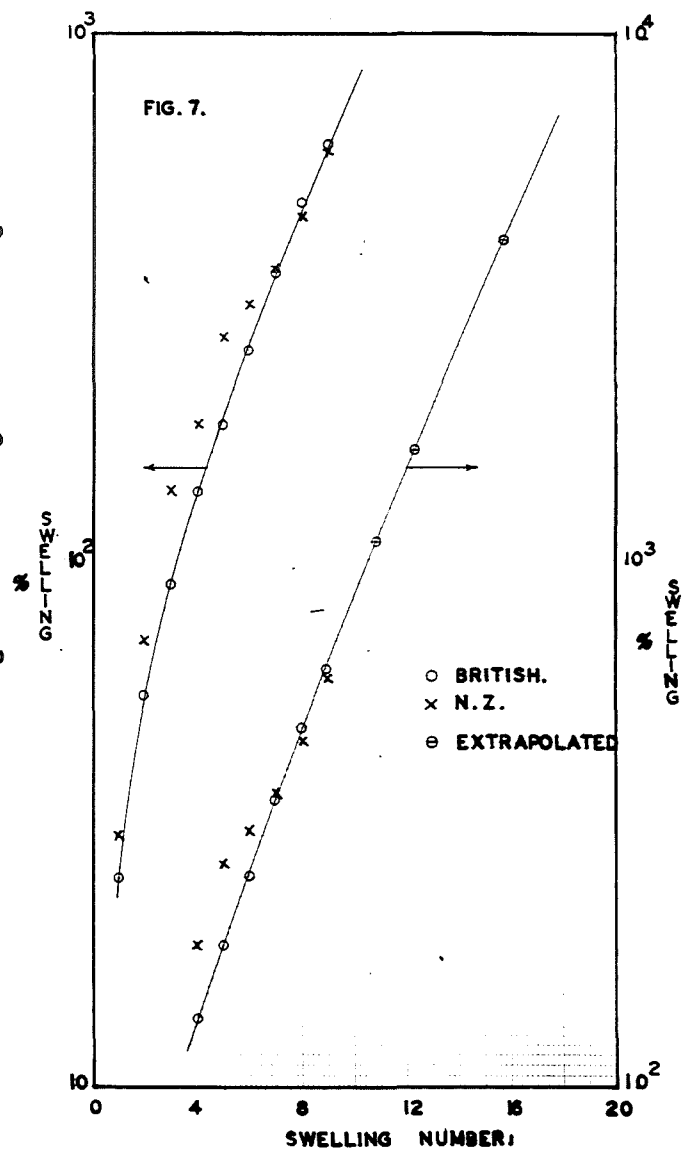
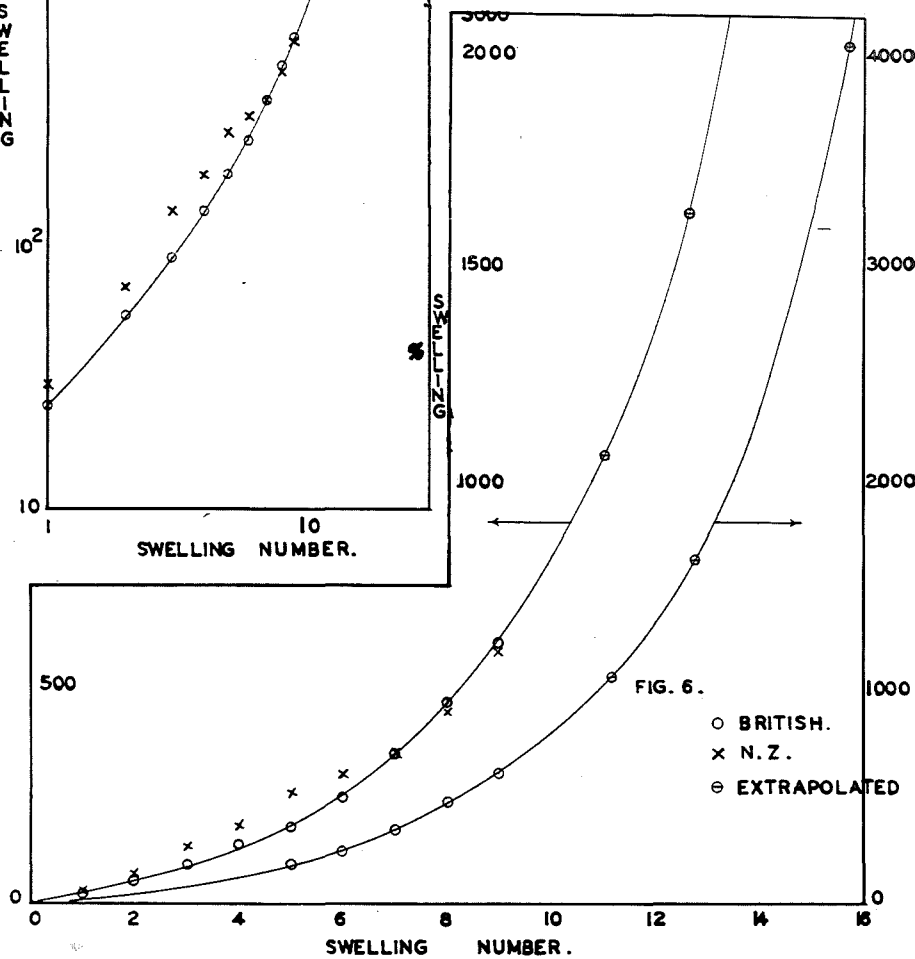
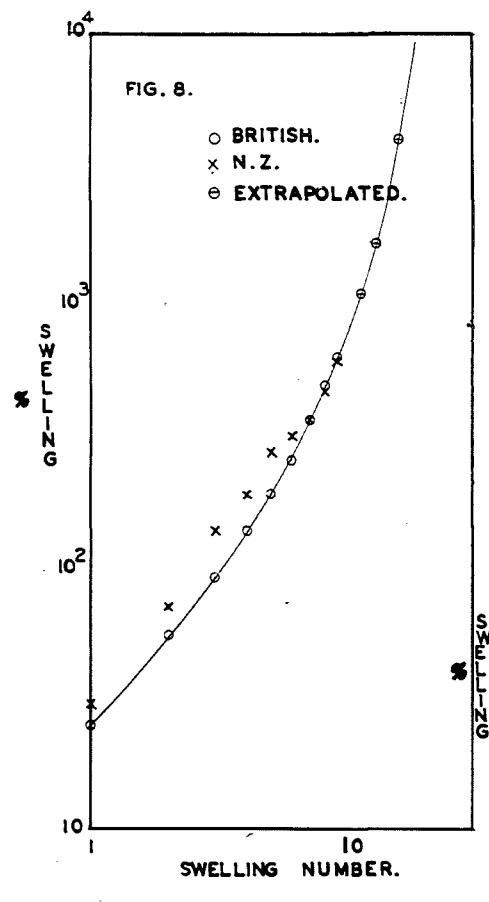
Table 1

Sw. No.	Percentage Swelling	
	British Coals	New Zealand Coals
1	25	30
2	55	70
3	90	135
4	135	185
5	185	265
6	260	305
7	350	355
8	475	450
9	615	595
9 <sup>+</sup>		1070
9 <sup>++</sup>		1630
9 <sup>+++</sup>		4050

Suggate (1) points out that the figures for percentage swelling for New Zealand coals given by Spooner's formula do not form a true progression, which in view of the method of derivation is to be expected. The departures from true progression merely reflect the changes in rate of development of swelling properties between the two coal bands previously noted, fig. 5. That the point of inflection in the curve occurs in the range of swelling numbers 6-8, see fig. 6, may indicate that the onset of free swelling restriction in the crucible test is much more significant for New Zealand coals than for the British coals on which the test was based.

This lack of true progression makes estimation of the values on a linearly extended swelling scale of the 9<sup>+</sup>, 9<sup>++</sup> and 9<sup>+++</sup> buttons difficult. Suggate (loc. cit.) quotes 10 $\frac{1}{2}$ , 11 $\frac{1}{2}$  and 13 respectively, but does not give his reasoning in arriving at these particular values. After the discontinuity in the values for New Zealand coals the values appear to





follow closely those for British coals. Fig. 6 shows the functional relation for the British coals extrapolated in the 9 region with the swelling number scale linearly extended. The values indicated by this extrapolation for  $9^+$ ,  $9^{++}$  and  $9^{+++}$  are 11,  $12\frac{1}{2}$  and  $15\frac{1}{2}$ .

Figs 7, 8, which show the data of table 1 plotted on log. natural and log. log. scales and again extrapolated, also predict these values and show that the progression has no simple exponential or power function form.

Swartzmann (8) shows the variation in volume with swelling number. His curve is exactly the same as that in fig. 6 but is displaced vertically by 25% swelling - the percentage swelling assumed by Spooner as giving agglomeration with no nett swelling.

Calishazer and Spooner (9) claim that the crucible swelling test can be used as a sensitive\* test for the degree of oxidation of a coal and that experimental figures show agreement with the formula

$$\text{Swelling No.} = A e^{-Bt}$$

However, they show that this applies only after decrease in swelling number with oxidation commences; for some coals swelling number increases to a maximum first, an effect also noted by Kreulen (15). This seems to indicate some critical oxygen content for maximum swelling as suggested by Blayden, et al, (16) who postulates that oxygen content is related to the amount of tar produced on carbonisation and that it is this tar which determines the degree of swelling. This effect could be related to the region of rapid decrease of swelling number with increasing carbon percentage composition seen in fig. 5.

Dalahunty and Harrison (10) and Berkowitz (17) show that the swelling of a coal is dependent on the rate of heating and thus on the rate of phase

\*This is not the opinion of many other workers however, e.g. Kreulen D.J.W. (loc. cit.).

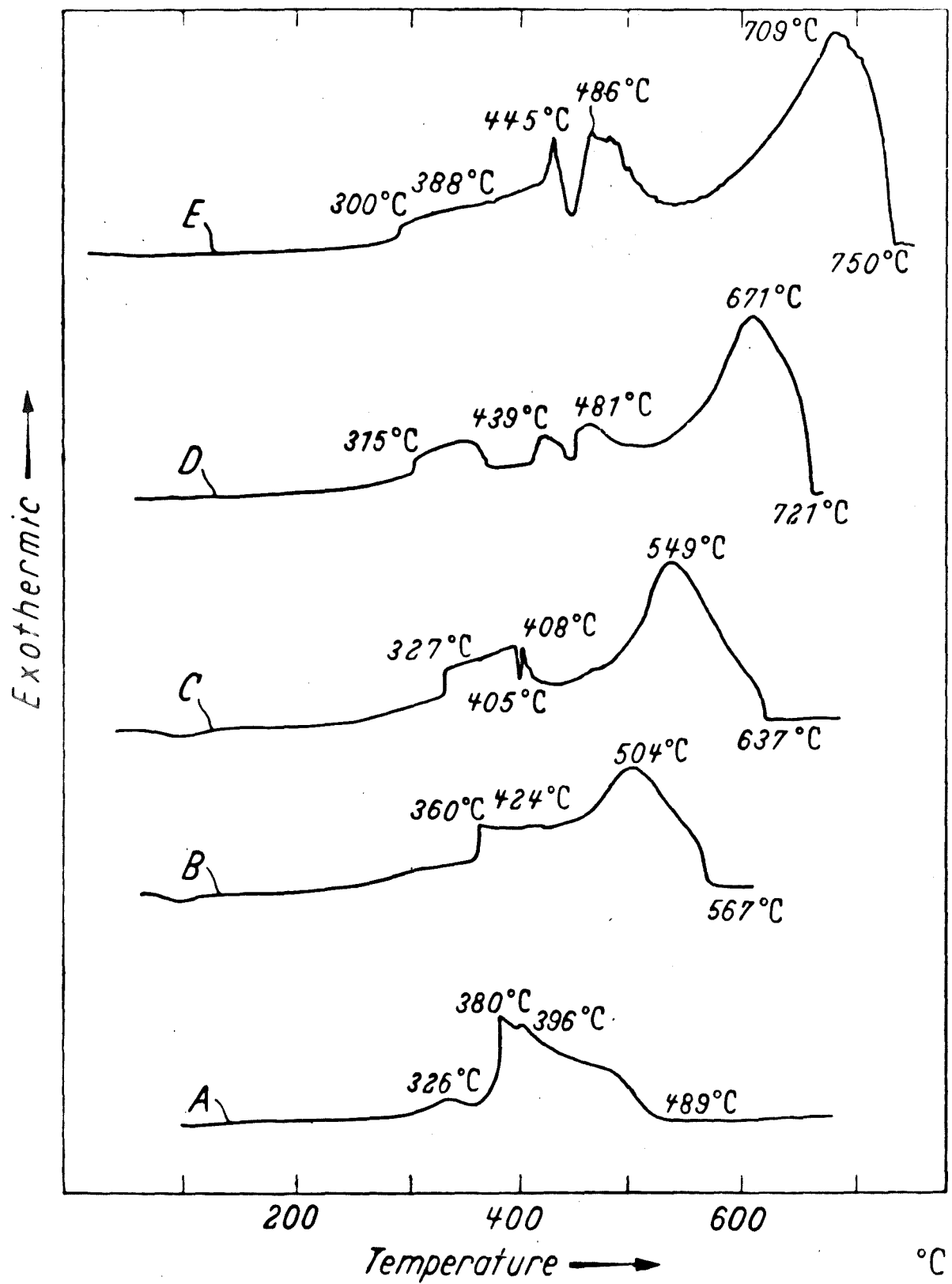


FIG. 19

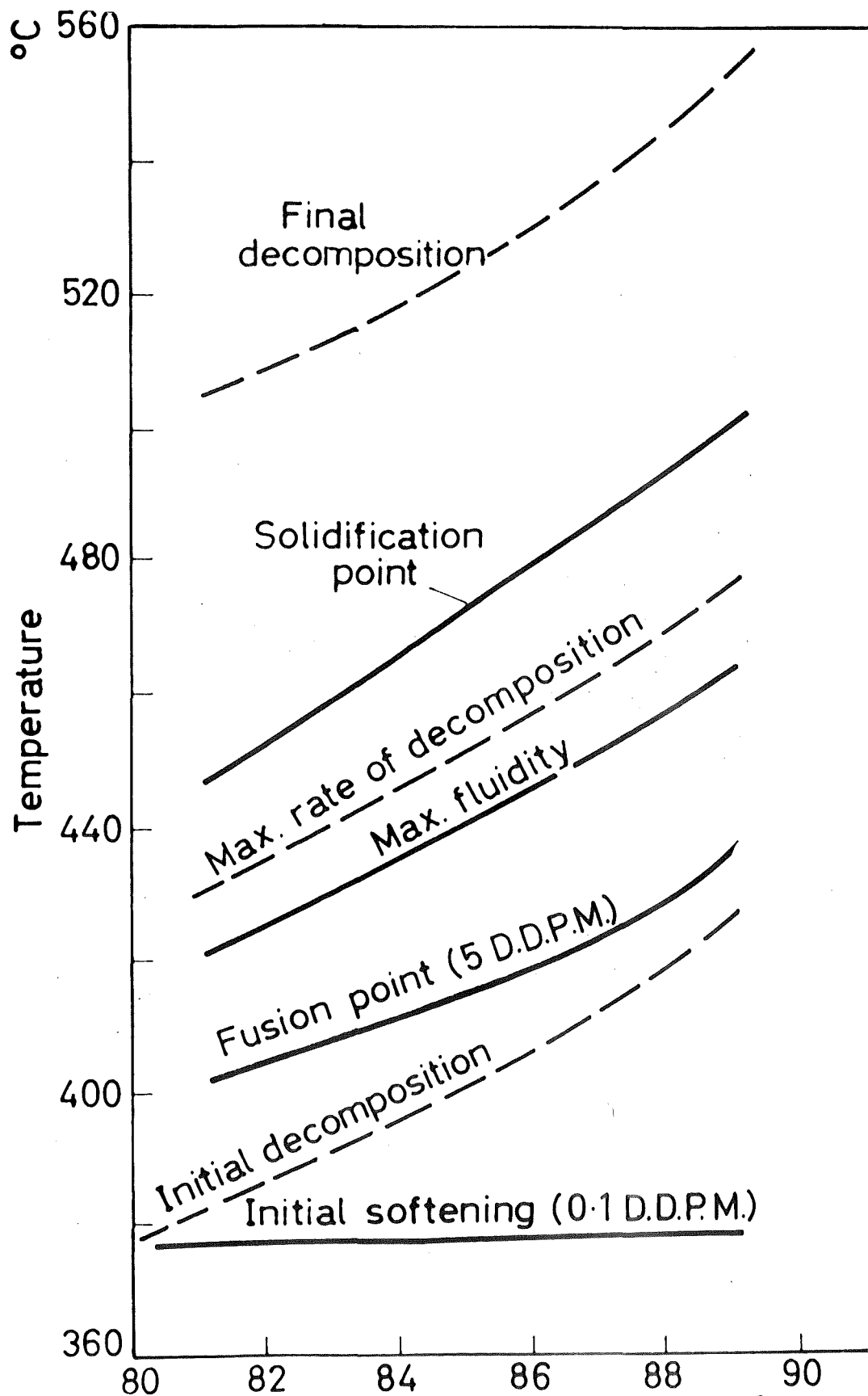


FIG 19 A Carbon content, per cent (d.a.f. basis)

transition and volatile evolution.

Macrae and Murthi (11) show that changes in fluidity occur without active decomposition taking place. They postulate the evolution of a plasticizing material to the surface of the particles, a froth formation of this material giving a rapid initial increase in fluidity followed by a constant rate of decrease of fluidity as the plasticizer foam is broken up allowing increasing mutual enmeshment of the remaining solid particles. These phenomena are shown as time dependent in tests made at constant temperature. They are emphatic in their assertion that no decomposition signified by weight loss occurs during these processes, but suggest a mild pyrolysis may be responsible for formation of the plasticizer.

Waters (12) shows that fluidity and decomposition are phase related phenomena for a coal heated to give a constant rate of temperature increase, and that they are rank dependent with respect to their characteristic temperatures at which they become evident, attain a maximum value and disappear. He shows also a "softening" region similar to the region in which Macrae and Murthi (*loc. cit*) worked. He shows this region to become more significant as the rank increases, fig. 19A, as softening itself is independent of rank. Waters also presents evidence in support of the hypothesis that such softening is a reversible physical change and does not represent any form of decomposition. Stott and Baker (13) working with New Zealand coals show, fig. 19, that the characteristic temperatures in the curve obtained for these coals during differential thermal analysis are rank dependent. Under these conditions, in effect controlled combustion, coals undergo a predominantly exothermic reaction.

Millard (14) shows in contrast that in carbonisation the coal undergoes endothermic reaction at temperatures below 700°C, that endothermicity is

a maximum at 600°C-650°C, and that at 700°C a dramatic change to exothermicity occurs. This effect is very likely partly responsible for the final temperature versus time characteristic obtained in the present apparatus.

Brown (22) in presenting a comprehensive survey of work done on property assessment for Australian coals shows that temperatures of initial and maximum fluidity and decomposition are phase related and rank dependent, decomposition occurring at higher temperatures than fluidity. This phase displacement becomes greater with increasing rank. Evidence is also given to show that decomposition of coals proceeds by discrete proportions as the temperature increases, i.e. that various components having a range of activation energies are involved. Brown also shows that the plastic layer in a carbonising coal charge is of the order of 3 mm in thickness.

This conclusion is challenged in discussion by Nadziakiewicz who states that the width of the plastic layer is a rank dependent property used in Poland as a classifying parameter for coking coals. He gives 7-30 mm as the range of widths encountered in his own experience of measuring this property. Nadziakiewicz and Calloott and Innes (23) both used a penetration plastometer in measuring the plastic layer width whereas Brown quenched a charge during carbonisation and then prepared cross-sectional micrographs, a more direct method. Brown's results indicate that softening, distinct from fluidity, occurs to varying degrees depending on rank and coking properties. It seems that this softening is indistinguishable from plasticity by penetration plastometer techniques. All these workers agree that the temperature gradient through the plastic layer is very steep.

Brown also shows that for higher rank coals the greater proportion of volatile evolution occurs at temperatures at which solidification of the coke has commenced.

FIG. 9.

APPARATUS ASSEMBLED FOR A RUN.— DIAGRAMATIC.

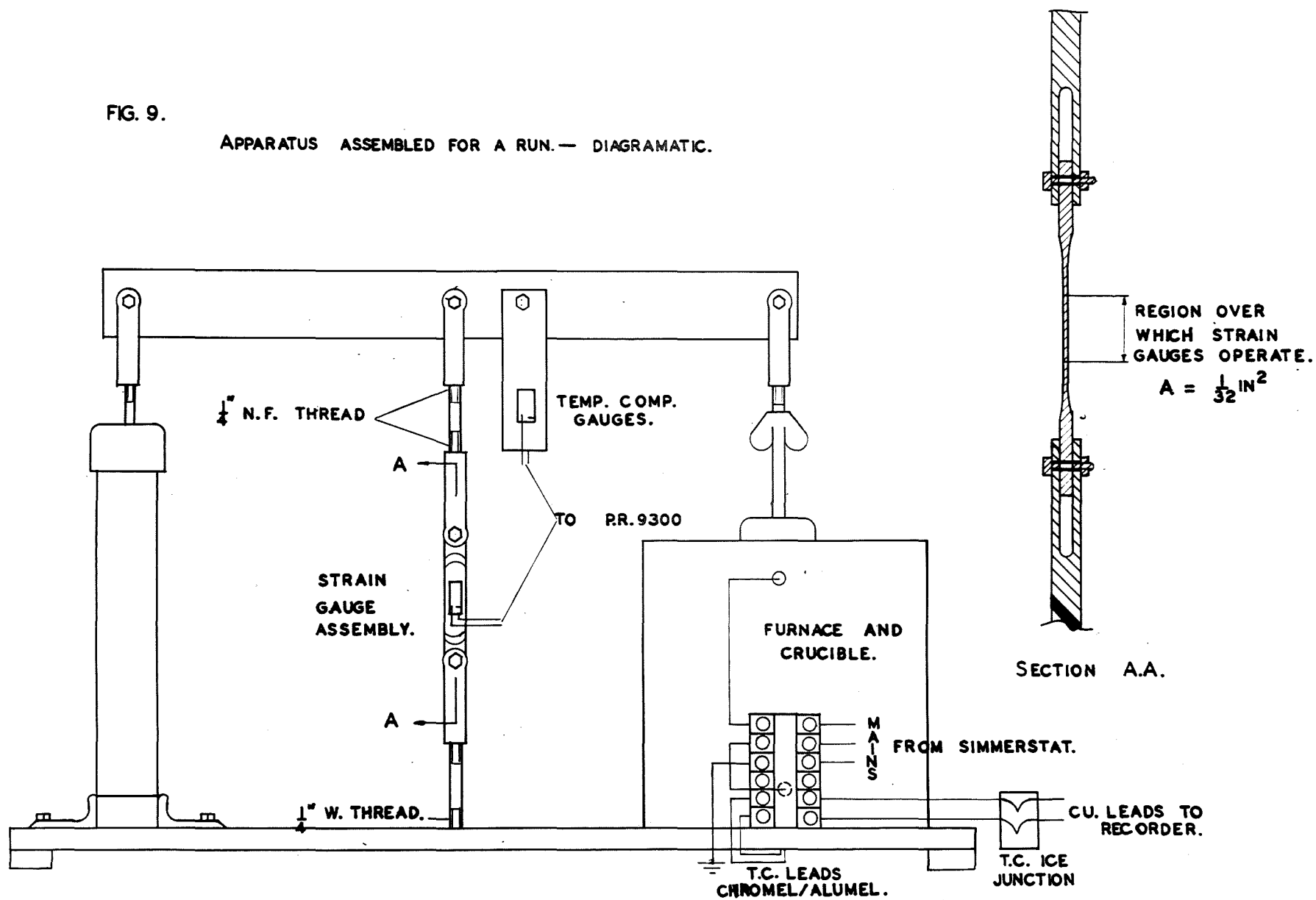
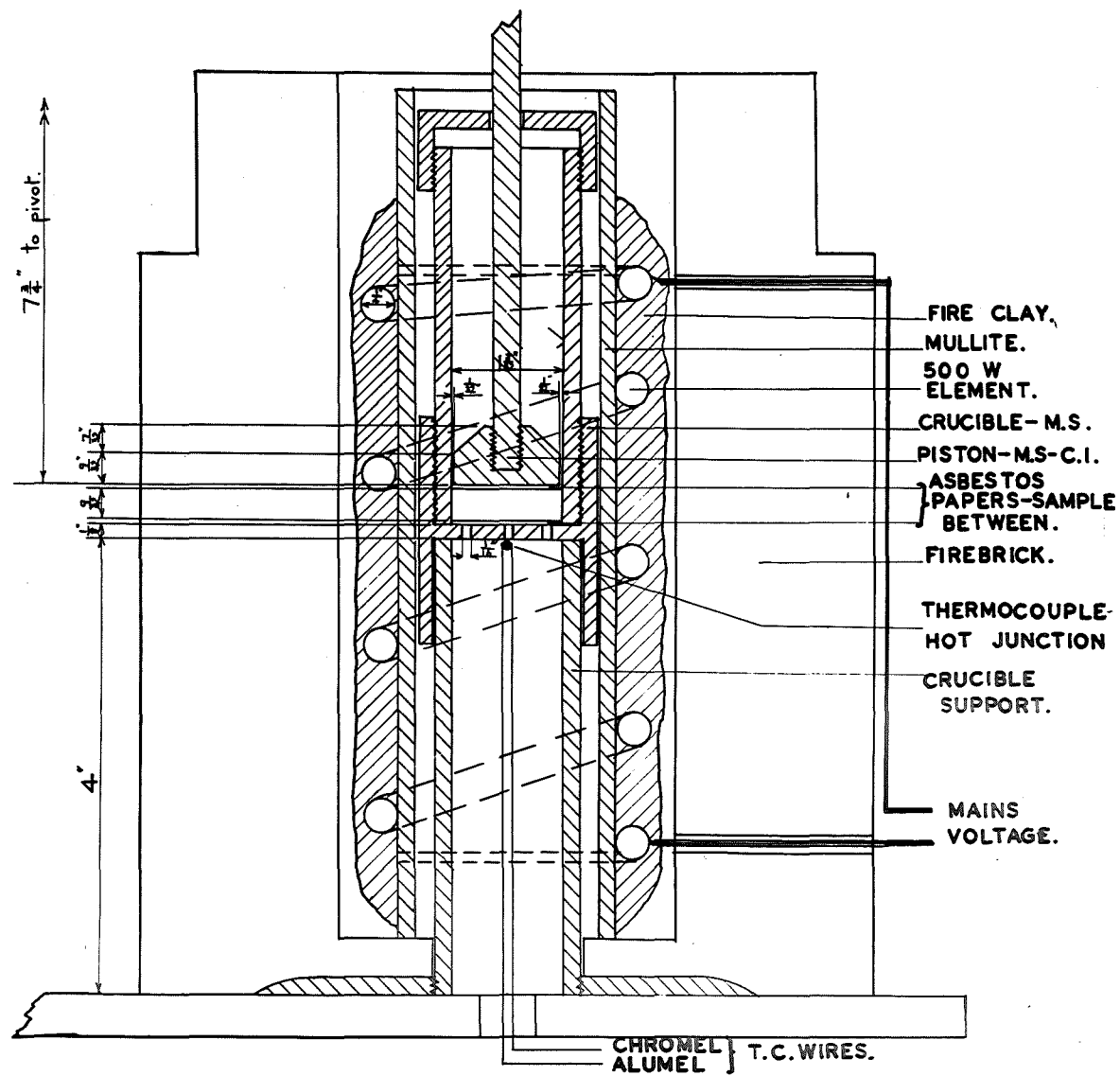


FIG 10. FURNACE AND CRUCIBLE  
DETAIL .





## III

APPARATUSModifications

## Pressure Measurement:

As discussed in the previous report on this work (4), the hydraulic pressure measuring device proved unsatisfactory. The difficulty was to obtain rigidity in the lever and pressure measuring system so that constant volume conditions in the crucible were assured. As an alternative to an hydraulic system it was decided to use a mechanical system with strain gauge detection of pressure. Philips paperback strain gauges, PR 9814, were used, for which  $R = 120 \text{ ohms} \pm 0.5\%$ ,  $K = 2.00 \pm 1.5\%$ ,  $\alpha_t = 32 \times 10^{-16} \text{ ohms/ohm/}^\circ\text{C}$ .

The gauges were mounted on a short piece of brass bar ( $\frac{1}{2} \times \frac{1}{8} \text{ in}^2$ ) reduced in section, A, over the length of the gauges to  $\frac{1}{32} \text{ in}^2$  in the manner shown in fig. 9. This cross-section was chosen after sensitivity considerations. The gauges were to be used in conjunction with a direct reading bridge, PR 9300, sensitivity,  $\epsilon_{\min}$ ,  $10^{-6}$  microstrains. A was determined from the relationship

$$\epsilon_{\min} = \frac{P_{\min}}{A} \cdot \frac{1}{E_{\text{brass}}}$$

$P_{\min}$  was taken to be 0.50 lbs which corresponds approximately to  $0.25 \text{ lbs/in}^2$  at the piston face. E for brass was taken as  $15 \times 10^6 \text{ lbs/in}^2$ .

This brass section was held between pivot pins in the yolks as shown, fig. 9. Gauges were attached to both sides of the brass to counteract any non-axial components of stress - the brass was machined to reduce its cross-section in such a way that non-axial stressing was a minimum, fig. 9.

As the gauges were to operate near the furnace, it was decided to incorporate a similar set of gauges on a non-stressed brass bar which was

hung from the main lever as shown. These gauges being at the same temperature as the active gauges gave temperature change compensation.

The electrical signal received from the gauges by the bridge was taken as a mV output signal to a Philips FR 4069 m/04 recorder. The strain gauge, bridge, recorder measuring device was calibrated by deadweight methods and was found to be linear in the range required, a bridge output signal of 5.72 mV representing 20 lb force at the piston face. As full-scale deflection over the 10 division recorder scale represented 10 mV, pressures in  $\text{lbs/in}^2$  at the piston face are given by

$$\text{p.s.i.} = \frac{\text{Recorder divisions} \times 10}{5.72 \cdot A_p}$$

where  $A_p$  is the piston face area. As  $A_p = 0.835 \text{ in}^2$  (piston diameter =  $1\frac{1}{32} \text{ in.}$ ) this gives

$$\text{p.s.i.} = 4.185 \times \text{recorder divisions.}$$

This conversion factor has been used in calculating results tabulated in table 2 from the recorded pressure traces for Runs 1-40.

#### Recording of Results:

In order to obtain continuous pressure and temperature traces on the same time scale a twelve-point recorder was used. By connecting alternate recording points in two parallel groups of six and operating the recorder at 40 mm/hr time scale a good trace of both variables was obtained.

However, the signal from the thermocouples had a range of 0-36 mV over the temperature range 0-800°C used. Thus to record this signal on the same instrument as the 0-10 mV pressure signal required the inclusion of a voltage divider in the thermocouple lead circuit. A 1 in 4 voltage divider using resistors of 33 ohms and 100 ohms was used to give a full-scale reading of 40 mV for the temperature trace. The recorded values

have been interpreted in  $^{\circ}\text{C}$  values using Leeds and Northrup thermocouple tables (7).

#### Temperature Control:

The final major modification to the apparatus described previously (4) has been the automatic generation of a linear temperature increase in the coal sample. This has been effected by the use of a "Simmerstat" on-off controller and a small synchronous electrical motor driving through one revolution in four hours. A temperature rate of increase of  $200^{\circ}\text{C}$  an hour was wanted.

Experiment showed that over the temperature range  $200^{\circ}\text{C}$ - $800^{\circ}\text{C}$  the heating coil showed no significant temperature coefficient of resistance, fig. 13. Also an experiment was carried out to determine the watts input versus equilibrium furnace temperature to determine the effects of radiation and convective heat loss from the furnace as the temperature increased. Table 2 gives these experimental values. For each point temperature readings were taken at 5 minute intervals and three successive readings equal  $\pm 0.5^{\circ}\text{C}$  were taken to indicate thermal equilibrium at that input. Excessive draught was found to influence the equilibrium maintained furnace temperature by up to  $10^{\circ}\text{C}$ , so care has been taken that the furnace has not been subjected to obvious draughts during operation. For this experimental work the supply to the heating element was through a variac.

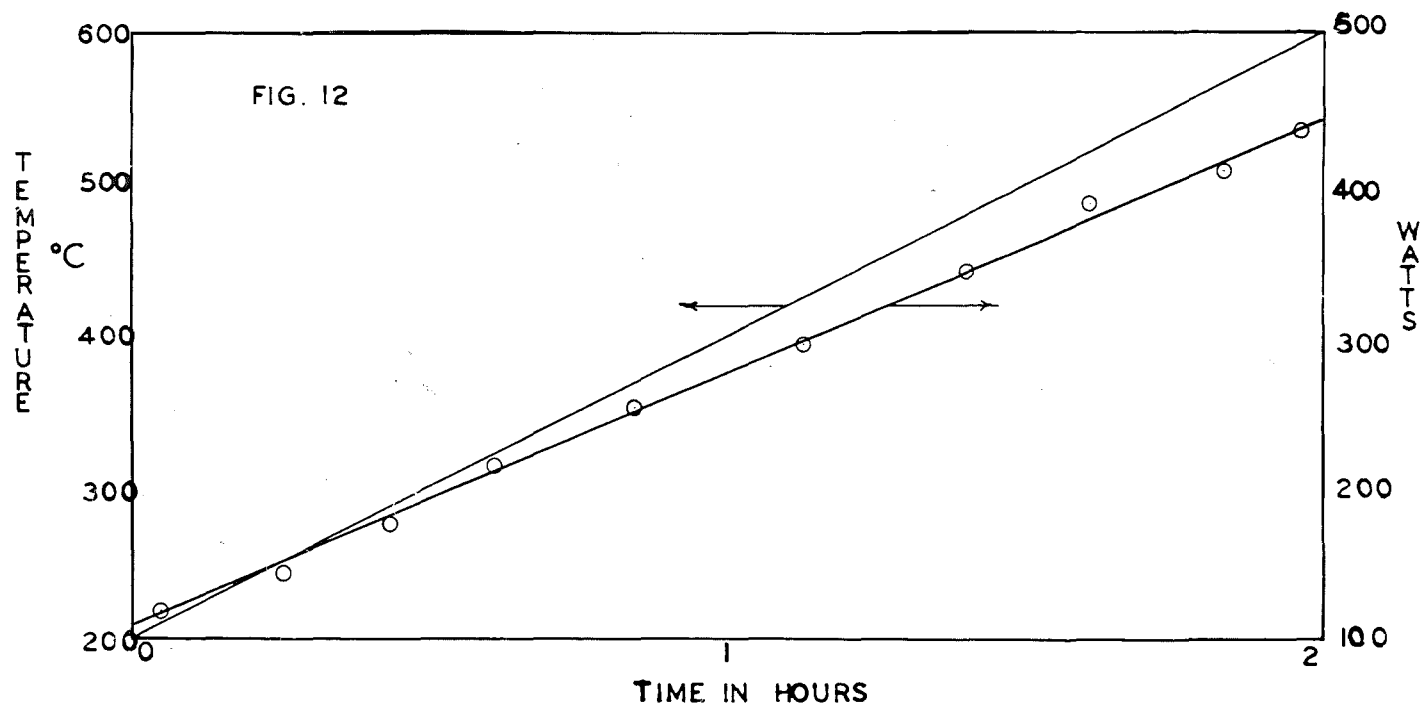
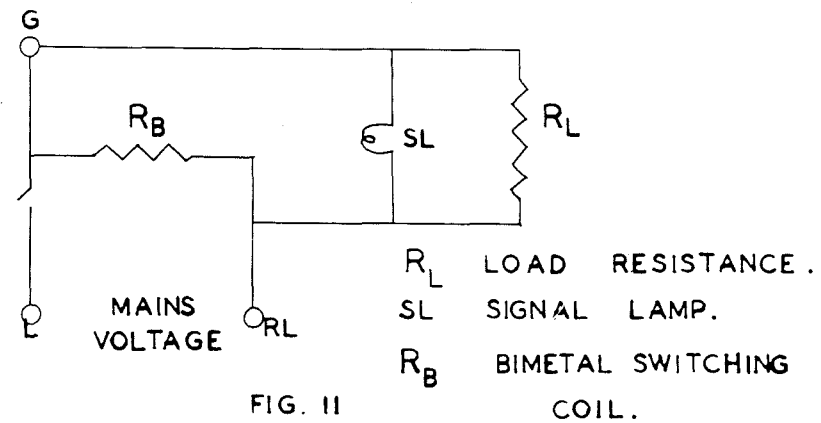
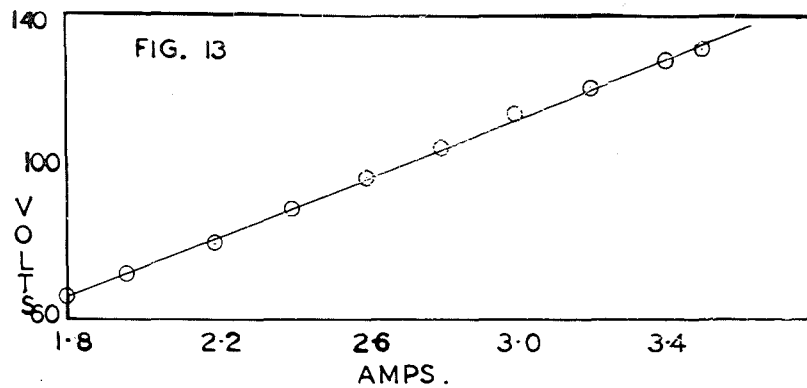


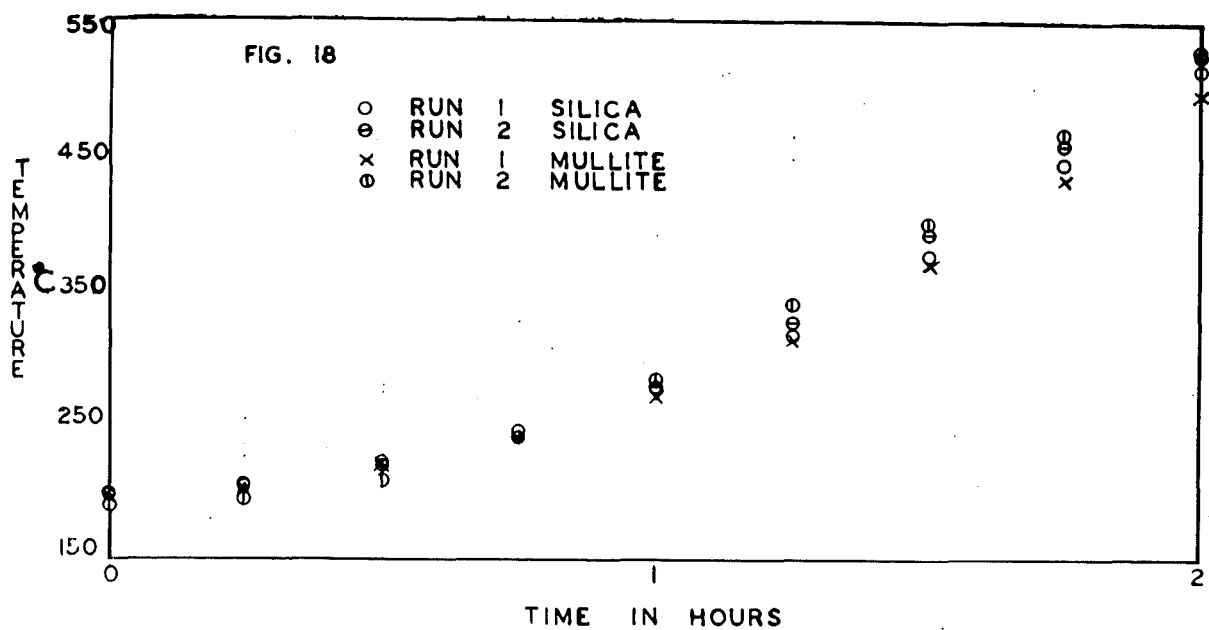
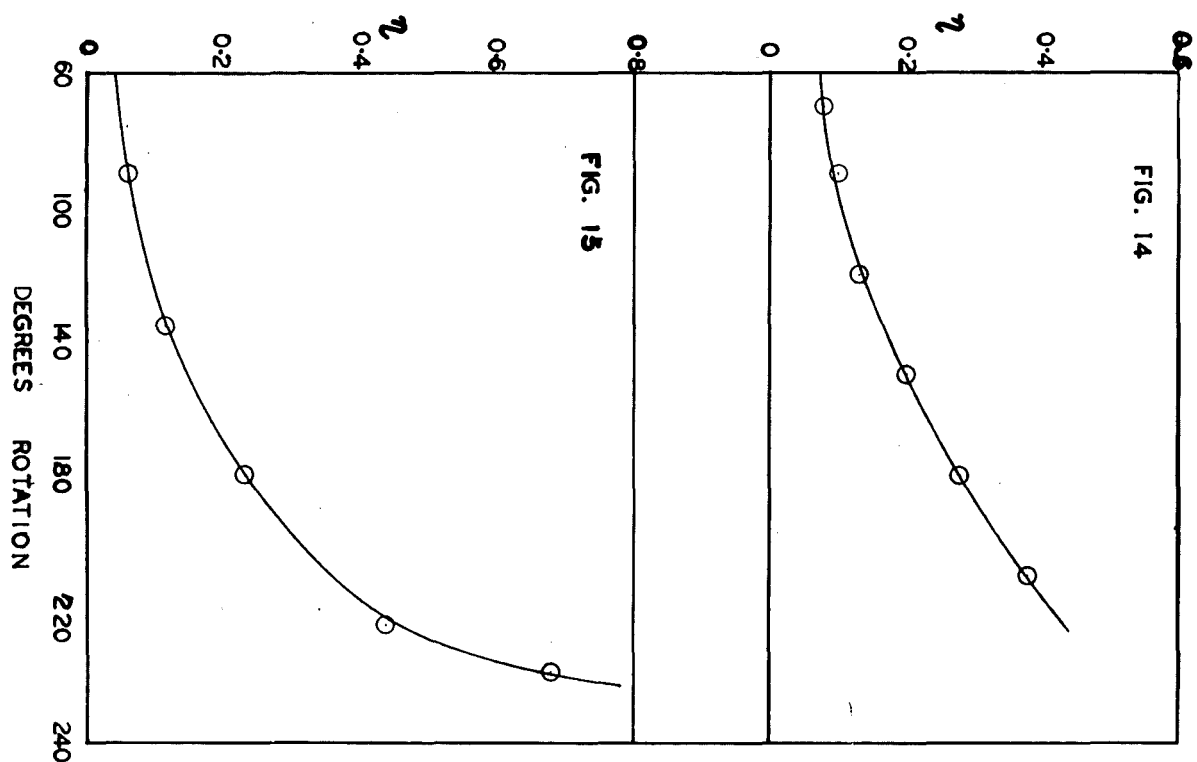
Table 2

Amps	Volts	Watts	Temp. °C
3.5	131	459	618
3.4	128	436	593
3.3	124	409	568
3.2	121	387	521
3.0	114	342	480
2.8	105	294	426
2.6	97	252	369
2.4	89	214	324
2.2	80	176	288
2.0	72	144	253
1.8	66	119	212

Fig. 12 shows the desired rate of increase of temperature with time, and from the figures of table 2 the required wattage input versus time curve to give this temperature versus time characteristic has been determined and is also shown. It is seen that it is substantially a linear relationship. In view of the difficulty in devising a drive for the variac to give the required input versus time characteristic the use of a Simmerstat on-off control, fig. 11, was investigated, the control to operate at a constant voltage and be driven through its operating range in such a way that the required wattage versus time characteristic was achieved.

For such a temperature control to be satisfactory, the following assumptions and requirements were necessary.

- (1) The time lag between heater input and furnace temperature must be constant over the range.
- (2) The discontinuous input from the switching of the on-off controller produces heating pulses which must be smoothed by the heat capacity of the furnace and crucible.



- (3) The fraction of total time,  $\eta$ , during which the controller is on must increase linearly with time.
- (4)  $\eta$  must have a sufficient range of values to give the required watts input range to the heater, i.e. at least a 1:6 ratio between extreme ends of the range.
- (5) The value of  $\eta$  must be reproducible at a given setting of the "Simmerstat" controller.

From the previous work, (1) appeared likely and so work was done to determine the validity and possible attainment of (2), (3), (4) and (5).

A degree scale was attached to the Simmerstat shaft with its  $0^\circ$  line at the positive off position of the controller and a pointer was attached to the spindle to move over this scale as the Simmerstat shaft rotated. It was found that no reliable switching occurred at a setting of less than  $60^\circ$  rotation from the positive off position.

Fig. 14 shows  $\eta$  versus rotation of the Simmerstat as obtained for tests with the constant voltage supplied through a variac. The characteristic was obviously nonlinear but did have a sufficient range of values and was reproducible  $\pm 2\%$ . However, induction effects from the variac produced sparking on switching which ultimately welded the Simmerstat points in the on position. It was also found that a voltage of 200 was required to give the required temperature range. For these reasons it was decided to try the Simmerstat control on mains voltage. Fig. 15 shows  $\eta$  versus rotation of the Simmerstat under these conditions.

The characteristic is still nonlinear but reproducibility and range requirements were found to be satisfied.

In all these tests requirement (2) was found to be met and so it was decided to use the Simmerstat, driving it through a cam device in such a

way that the linearity requirement was met.

To give the required range of  $\eta$  corresponding to the required rate of temperature increase, it was necessary to drive the Simmerstat through  $150^{\circ}$  in two hours. A small synchronous motor driving through  $360^{\circ}$  in four hours was available and this was used through a suitable reduction gearing to drive the Simmerstat in several test runs to determine the temperature versus time characteristic resulting from the combination of factors (1)-(5) under actual test run conditions. For these runs the furnace and charged crucible were soaked at  $200^{\circ}\text{C}$  with the Simmerstat set at  $71^{\circ}$  and then the motor drive was started and the temperature recorded at quarter-hour intervals. The recorded values for the three runs as shown in table 3 are plotted in fig. 16.

Table 3

Time from Start hrs	Temperature $^{\circ}\text{C}$		
	Run 1	Run 2	Run 3
0	192	196	203
$\frac{1}{4}$	226	233	252
$\frac{1}{2}$	273	278	302
$\frac{3}{4}$	326	333	356
1	381	390	418
$1\frac{1}{4}$	449	464	498
$1\frac{1}{2}$	528	545	568
$1\frac{3}{4}$	621	645	
2			

Fig. 16 shows:

- (1) agreement between runs,
- (2) the degree of nonlinearity in the temperature versus time curve,
- (3) that the average rate of increase of temperature is  $10^{\circ}\text{C}$  in 3 minutes, i.e.  $200^{\circ}\text{C}$  in an hour, as required.



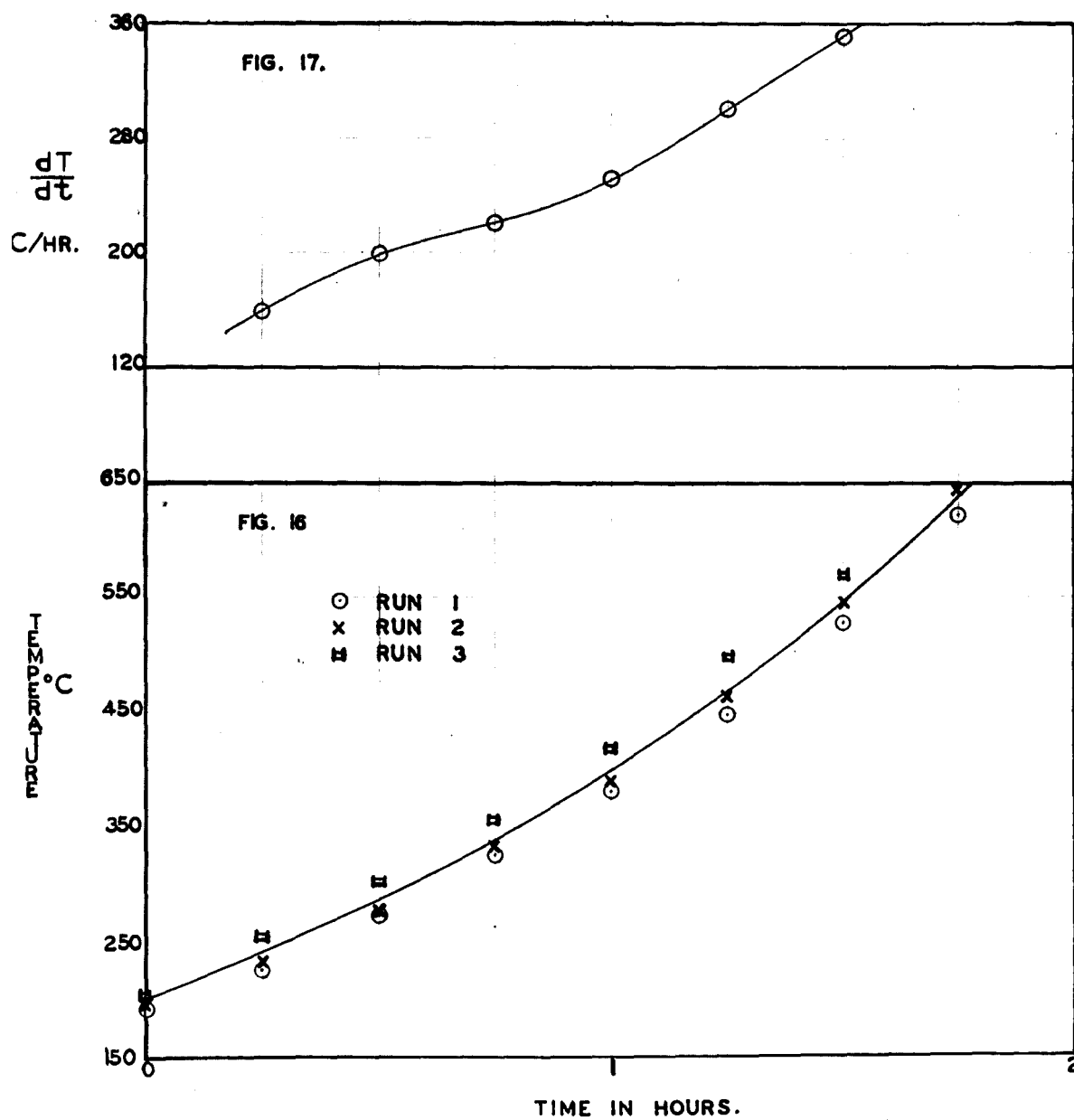


Fig. 17 shows the rate of increase versus time curve from which the cam profile required could be calculated.

In view of the value of the average rate of temperature increase and the relatively minor departures from the required  $200^{\circ}\text{C}/\text{hour}$  until high temperatures are reached, it was decided to use the apparatus with the motor drive used in these test runs. As long as the shape of the characteristic temperature versus time was reproducible the runs would give comparable pressure development in the coal samples and departures from the average  $10^{\circ}\text{C}/3 \text{ mins}$  value were small enough to have little effect on the pressure versus temperature curve, and on absolute pressure values at a particular temperature. The recorded results of runs 1-40 show that the temperature versus time characteristic was sufficiently reproducible for the purposes of the work carried out. The various factors affecting the rate of heating combine to give a characteristic which is almost linear over the temperature range used.

#### Lever System:

The rigidity of the lever system used is important in that flexing in it will cause friction in the pivots and correspondingly low recorded developed pressures and excessive deflection would mean that the samples were not kept at constant volume. For these reasons a heavier lever,  $1 \times \frac{1}{4}$  mild steel, has been used. For this lever deflection  $y$  is  $\frac{1}{500}$  in. for 20 lbs force on the piston face and  $\frac{L}{I} = 1000$  which is a satisfactory value to give no flexing.

#### Element:

Initially the heating element was wound on a fused silica former and

covered with fire clay. It was found that after repeated heating and cooling in runs this silica failed. Inspection of the failure showed some crystalline formation to have occurred due to the heating effect where the element was in contact with the silica and this had been the point from which the failure had proceeded. The silica was replaced in subsequent elements by a mullite tube which gave no trouble throughout the runs made. This did necessitate turning down the flange and cap of the crucible as the internal diameter of the mullite tube was  $1\frac{1}{2}$  ins. This meant the crucible fitted into the element much more closely and test runs were made to determine if any major change in the temperature versus time characteristic had occurred because of the new arrangement. Fig. 18 shows two runs with the silica tubed element and two with the mullite tubed element. Variations between duplicate runs and elements are due only to differences in starting temperatures and experimental error and are not significant of any real change in the characteristic shape of the curve. The data for fig. 18 is given in table 4.

Table 4

Time from Start hrs	Temperatures °C			
	Silica Element		Mullite Element	
	Run 1	Run 2	Run 1	Run 2
0	200	199	198	190
$\frac{1}{4}$	206	205	204	194
$\frac{1}{2}$	221	218	217	208
$\frac{3}{4}$	244	240	240	240
1	277	278	270	284
$1\frac{1}{4}$	317	326	314	340
$1\frac{1}{2}$	376	391	370	400
$1\frac{3}{4}$	444	459	434	467
2	514	527	497	530
$2\frac{1}{4}$	604	618	582	
$2\frac{1}{2}$	705	719	683	

### Sample Temperature:

One further factor which has been considered in specifying the apparatus has been the known effect on pressure development of the direction of travel of the temperature profile in a coal sample during carbonisation. As discussed previously (4) the aim in this type of apparatus using a small metal enclosed sample is to eliminate this effect, ie. to consider the sample to be a unitemperate mass. In this way the pressures measured become significant of the changing properties of a small volume of coal in a bulk being carbonised as it passes through the various stages of the carbonisation reactions, and the apparatus provides fundamental information on these changes as well as a more quantitative swelling property test. To obtain an indication of the degree of attainment of unitemperate conditions two runs were made in which a second thermocouple was placed in the centre of the piston bearing on top of the sample. The temperatures at this point and at the usual temperature measuring point, fig. 10, are recorded in RUN 35 and RUN 36. An analytical attempt to establish the temperature profile in the sample is given in appendix 1 and the results obtained are discussed in a later section.

### Specification

Figs 9 and 10 show the apparatus diagrammatically with detail of the strain gauge assembly and the furnace and crucible construction.

Plate 1 shows the apparatus assembled during a test run.

For the apparatus to be of any use as a standard test with the results presented as a calibration, the dimensions given would need to be rigidly adhered to and the test procedure detailed in the next section would have

to be closely followed. Under such conditions the apparatus gives results reproducible to  $\pm 5\%$  which compares favourable with those obtained by overseas workers with similar apparatus.

The element is of 500 watts. The wire is wound into a  $\frac{1}{4}$  in. diam. spiral, which is then wrapped around the  $7\frac{1}{2}$  in. long mullite former as shown in fig. 10, the turns being equally spaced. The element is held in place by insulating fire-clay which also evens out the heating effect over the length of the element. The finished element is approximately  $2\frac{3}{4}$  in. diam. and fits into the 3 in. diam. centre drilling in the fire-brick furnace. It rests at its bottom end on a "toe" of fire-brick. The crucible and its support are made of 1 in. diam. steam pipe and fittings. The bottom flange of the crucible is turned out to make it fit snugly, but easily, over the support. The  $\frac{1}{8}$  in. base plate has 13  $\frac{1}{16}$  in. diam. holes drilled in it to allow gas escape. These are equally spaced over the base. The flange and cap are turned down so that the crucible slides into the  $1\frac{1}{2}$  in. I.D. mullite tube readily. The cast iron piston is  $1\frac{1}{32}$  in. diam. has  $\frac{1}{16}$  in. clearance on its diameter in the turned bore of the crucible and the mild steel piston rod has a similar clearance in the crucible cap.

These clearances and component fittings in the crucible-furnace construction are important as some oxidation does occur and difficulty results from too close fitting components especially when runs continue to  $800^{\circ}\text{C}$  and above. It is also important that the crucible rests rigidly on its support and does not tilt or bed down when pressure is applied as this would introduce zero errors and excessive piston friction. The piston clearance also allows gas escape. The thermocouple hot junction is positioned just touching the centre of the crucible base.

The chromel and alumel wires, fused at the hot junction, go out through the crucible support to the panel shown in fig. 9. They are

insulated over their complete length with porcelain fish spine beads and held in place by an asbestos packing in the crucible support. Each wire is joined to a copper lead in the cold junction - a dewar vacuum flask. The element is heated using intermittent mains voltage supplied through the Simmerstat, as discussed previously, to the panel, and from there through fish spine bead insulated wires, which pass through the furnace wall, to the element.

The pivot positions on the lever arm are equally spaced drilled holes in which the pivot pins are a close fit. The pivoting action at all yokes occurs in the two yoke tines, in which the pins are a machined fit - these yokes are pivot members off aircraft controls. Adjustment of the lever system, required both for levelling the lever arm and pressure zeroing before a run, is provided by the threaded rods in the yokes. A thumbscrew was attached to the piston rod for levelling the main lever. Zeroing before a run, discussed in the next section, is carried out using the short rod at the bottom of the strain gauge assembly. It has a  $\frac{1}{4}$  in. N.F. thread into the yoke and a  $\frac{1}{4}$  in. W. thread into the base plate. Screwing it results in a nett shortening of the assembly.

The leads from the strain gauges and from the thermocouple cold junction to the recorder are of copper and their resistance has negligible effect on the pressure and temperature values recorded. Those to the strain gauges were soldered to the gauge contacts after the gauges were fixed on the brass, and the gauges and lead junctions were fixed using "Araldite". The fragility of the gauge contacts made this necessary.

The Philips PR 9300 bridge in the strain gauge circuit and the 1 to 4 voltage divider in the thermocouple circuit translate the milli-voltage signals from the detecting devices into signals of suitable magnitude that

satisfactory recording of both on the same recorder, PR 4069, is possible.

The temperature compensating strain gauges are fixed to brass in the same way as the active gauges and held in a similar position relative to the furnace. Thermal expansion effects in the two brass strips are then similar and connections to the gauges are made in such a way that these effects cancel each other out.

## IV

TEST PROCEDURE

As discussed in the previous report (4) on devising a suitable test apparatus there are several factors which can influence the swelling pressures developed in an apparatus of the type used. These factors, bulk density, particle size, rate of heating and sample age, must therefore be carefully controlled if duplicate runs are to be truly comparable and if runs on different coals are to give meaningful relative property indication. To ensure that the results obtained embodied these two conditions the procedure for each run followed a standard pattern.

To eliminate sample age effects a +7 mesh sample of each coal to be tested was ground and the -52 +72 mesh and -72 mesh fractions separated immediately before a run was made. Duplicate runs were then made in both the swelling pressure apparatus and the Standard Crucible Swelling Test apparatus. (This latter was made to the specifications given in BSS 1016 (2) and the assembled apparatus is shown in plate 2.)

The swelling pressure runs were made using a 3 gm sample of the -52 +72 mesh fraction. This sample was compressed to  $50 \text{ lb ft}^{-3}$  between asbestos paper as shown in fig. 10. A gauge was used to indicate the correct volume of the sample to give this bulk density and compression was effected by sharp light blows on a close-fitting brass piston bearing on the upper asbestos paper.

Prior to this sample preparation and crucible charging, the strain gauge bridge was always switched on to allow it to warm up as in the first two hours of running it had been found to suffer excessive drift. The charged crucible was then placed in position and the lever system connected and adjusted using the thumbscrew on the piston rod until the lever arm



was horizontal. The power was then switched on to the element and the Simmerstat set at the position known to give a  $200^{\circ}\text{C}$  furnace temperature with the driving motor switched out. As this  $200^{\circ}\text{C}$  temperature was approached the recorder was switched on and when a constant temperature trace was shown the run was commenced. During this pre-run period the strain gauges had no tension on them but merely hung freely from the lever arm. It was important that this pre-run period was long enough for complete thermal saturation of the crucible, sample and furnace at  $200^{\circ}\text{C}$  if reproducible temperature increase curves were to be obtained from run to run.

When all these conditions were satisfied and the run was to commence, the Simmerstat driving motor was switched in and the lever system made rigid at zero strain gauge pressure indication. This was done by connecting in the strain gauge assembly and adjusting by means of the bottom adjustment, fig. 9, until the recorder indicated correct zeroing. The apparatus was then left alone until the run terminated, i.e. zero pressure, or  $800^{\circ}\text{C}$  temperature indication occurred.

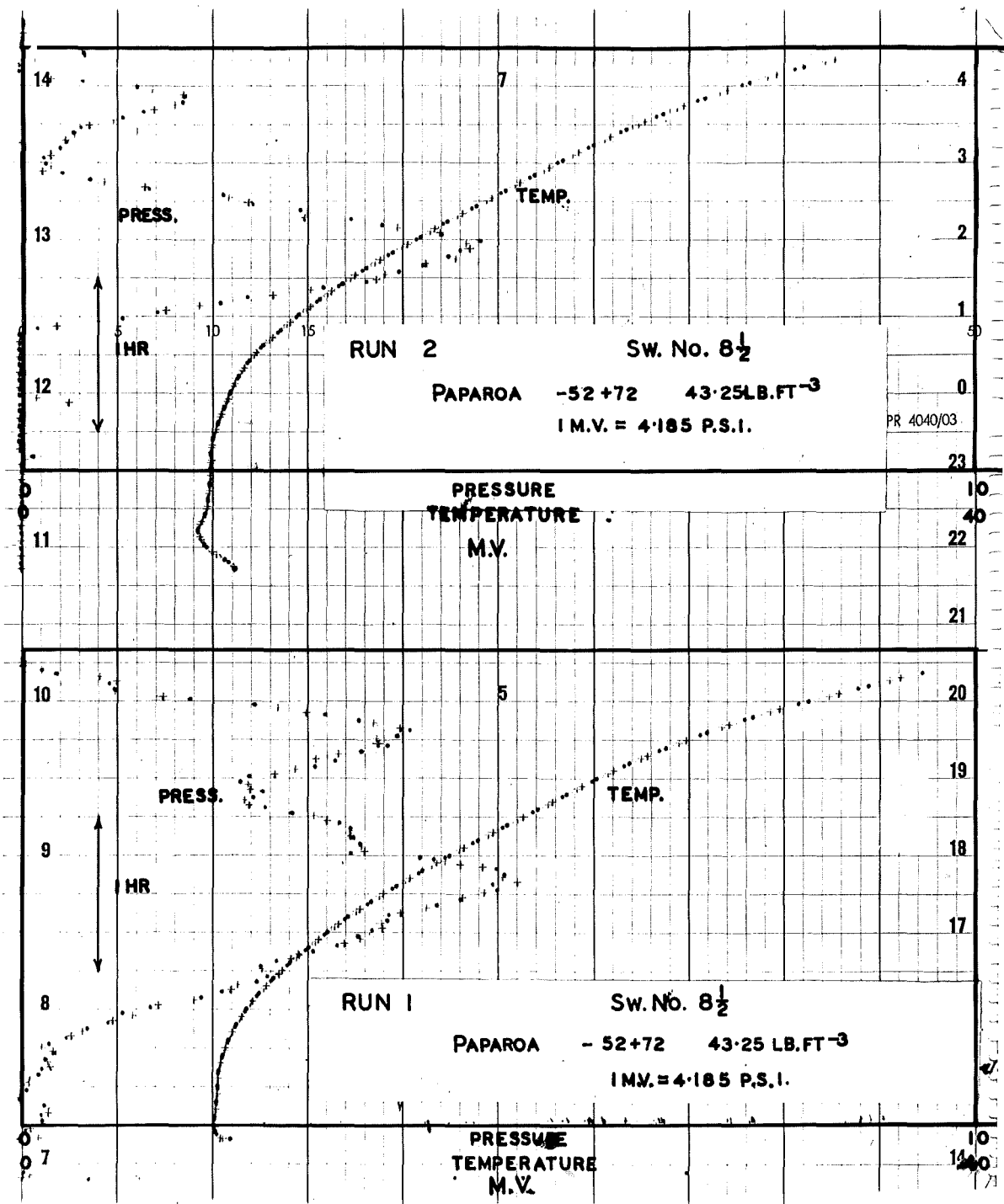
It had been previously determined that the temperature rise characteristic with respect to time was reproducible and so the condition of reproducible rates of heating from run to run was fulfilled. For the purposes of the work done reproducibility was the only criterion as no account of the effect of rate of heating was sought.

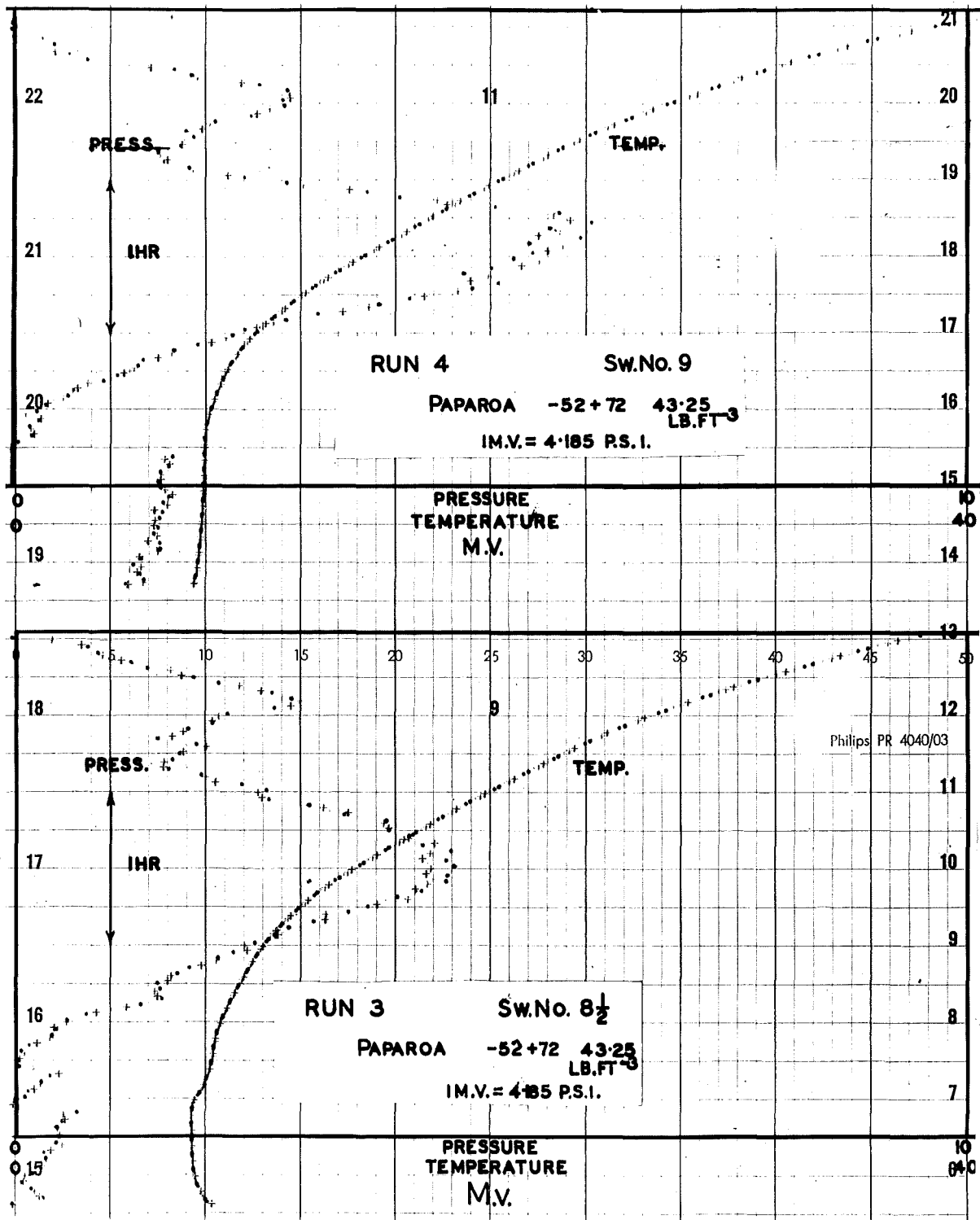
TABLE 5

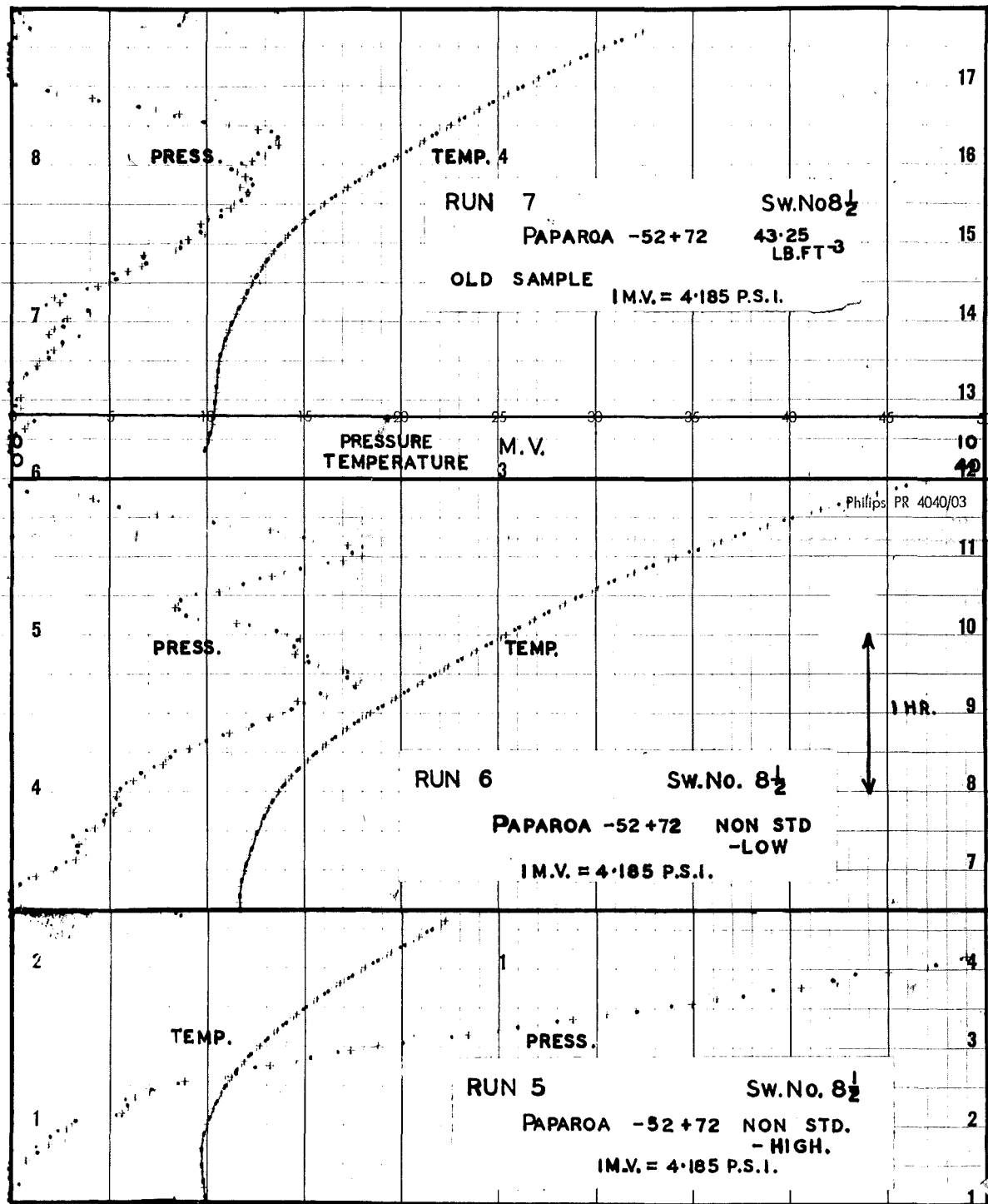
Run No.	Orig. Temp.	Char. Temp.	Char. P. psi	Corr. P. psi	Actual P <sub>max</sub> psi	Temp. at P <sub>max</sub>	Sr. No.	Coal	Comments
1	201°C	391°C	21.33	-11.3	0.14	230°C	8½	Paparon	Second peak well formed at 702°C.
2	195°C	400°C	20.05	-15.0	0.0	230°C	8½	Paparon	" " " at 692°C.
3	200°C	353°C	19.43	-6.95	1.05	223°C	8½	Paparon	" " " at 683°C.
4	197°C	409°C	25.25	-11.0	1.02	249°C	9	Paparon	" " " at 683°C.
5)	Show variations due to bulk density of sample - qualitative only						8½	Paparon	Higher than std bulk density-higher P developed.
6)							8½	Paparon	Lower than std bulk density-lower P developed.
7)							8½	Paparon	Low P - due to old sample. Std bulk density.
8							8½	Paparon	Power failure during run.
9	214°C	319°C	9.61	-8.35	Steady decrease from 214°C		1½	Strongman	Second peak tendency 628°C, op. 13.
10	217°C	313°C	12.12	-4.30	2.96	244°C	2	Strongman	
11	177°C	290°C	11.91	-7.40	3.73	207°C	1½	Strongman	
12	206°C	300°C	5.85	-10.22	Steady decrease from 206°C		1½	Strongman	Not comparable as -72 mesh sample.
13	206°C	310°C	11.50	-6.30	2.14	227°C	2	Strongman	Second peak tendency 615°C, op. 9.
14	192°C	295°C	13.17	-4.45	2.90	206°C	2	Blackball	Note exact reproduction of trace details 14 and 15
15	208°C	351°C	12.96	-11.5	2.66	224°C	2	Blackball	
16	191°C	329°C	11.29	-12.3	0.29	207°C	2	Blackball	End of increasing sample age.
17	204°C	334°C	19.64	-2.60	3.44	256°C	8½	Westport	Small second peak at 665°C (5.14 psi)
18	193°C	295°C	16.10	-1.35	3.34	231°C	8½	Westport	Small second peak at 630°C (5.14 psi)
19	200°C	334°C	14.22	-8.68	1.72	244°C	8½	Westport	Second peak at 650°C (8.7 psi)
20	192°C	314°C	13.12	-7.73	1.40	217°C	8½	Westport	Second peak not evident.
21	207°C	334°C	13.60	-8.10	1.89	272°C	8½	Westport	Second peak about 600°C. Zero setting of bridge in doubt.
22	194°C	335°C	19.84	-3.45	-0.50*	272°C	8½	Westport	New sample. Second peak 650°C (11.7 psi)
23	192°C	331°C	20.45	-3.35	-0.00*	276°C	8½	Westport	Mechanical effect finished run at 350°C.
24	192°C	324°C	16.75	-5.80	2.74	232°C	7	Fly Creek	
25	233°C	350°C	14.43	-7.37	Steady decrease from 233°C		7	Fly Creek	
26	188°C	315°C	15.28	-6.42	1.10	225°C	7	Fly Creek	
27	206°C	315°C	15.70	-2.95	3.00	245°C	7	Fly Creek	
28	199°C	324°C	19.65	-0.75	0.51	213°C	6	Mixed Sample	Second peak at 580°C (15.9 psi)
29	206°C	329°C	17.96	-3.60	0.85	226°C	6	Mixed Sample	Second peak at 600°C (10.9 psi)
30)									Runs to determine the thermal expansion effects in the apparatus.
31)									
32)									
33	199°C	319°C	20.25	0.20	1.31	240°C	6½	Mixed Sample	
34	189°C	329°C	13.80	-10.15	0.13	204°C	4½	Mixed Sample	Second peak at 710°C (7.0 psi)
35)									Runs to determine temperature profile in sample.
36)									
37	217°C	462°C	37.20	-4.70	7.85	317°C	9**	Paparon	Plateau actual pressure > 7.5 psi 280°C - 335°C
38	197°C	453°C	35.85	-7.95	7.20	346°C	9**	Paparon	Similar plateau > 7.0 psi
39									Bridge drift indication.
40	209°C	387°C	24.95	-5.50	Negative pressure 9** from start.		9**	Paparon	Long warm-up period.

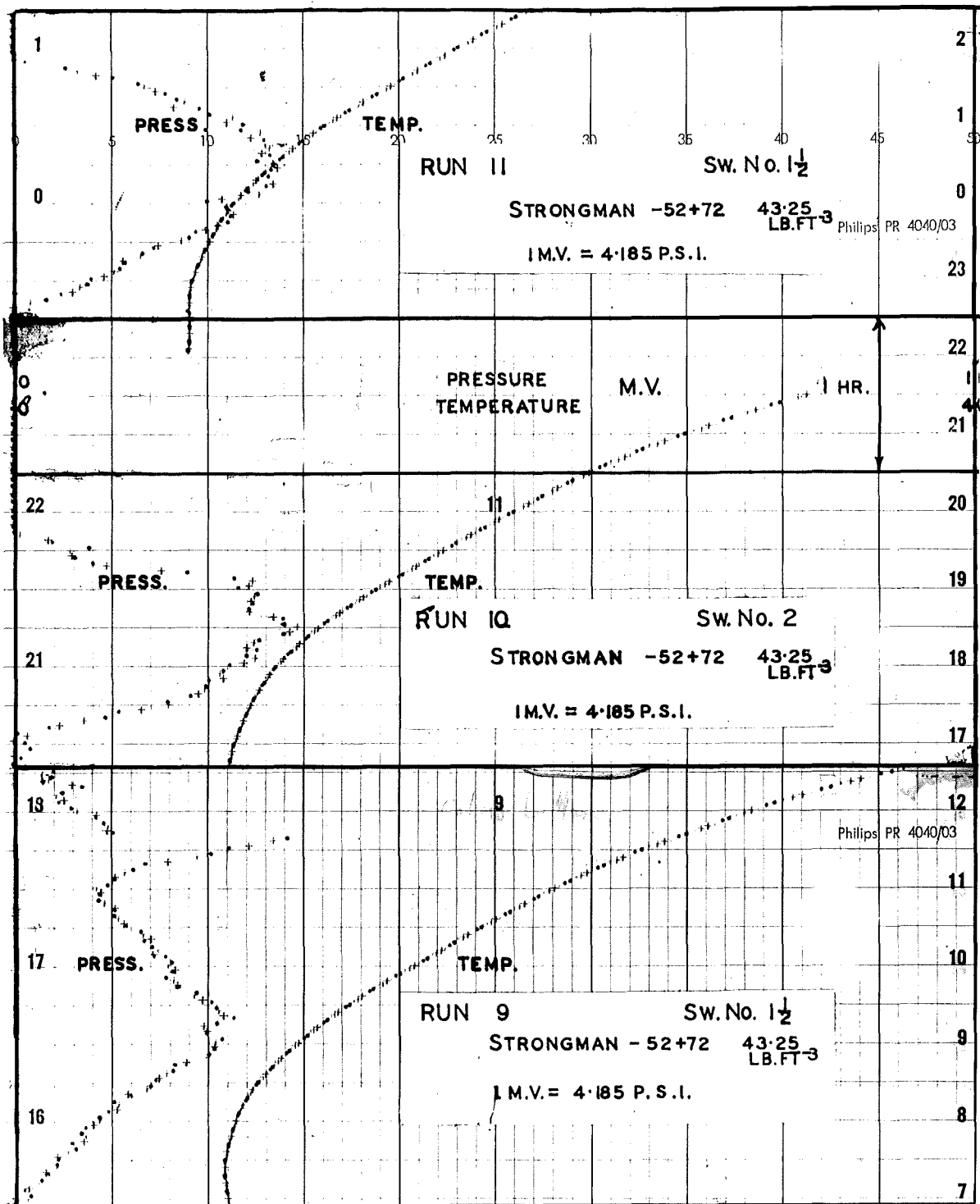
\* These low actual pressure peaks are probably due to "stickiness" of the piston at low temperatures due to insufficient clearance.

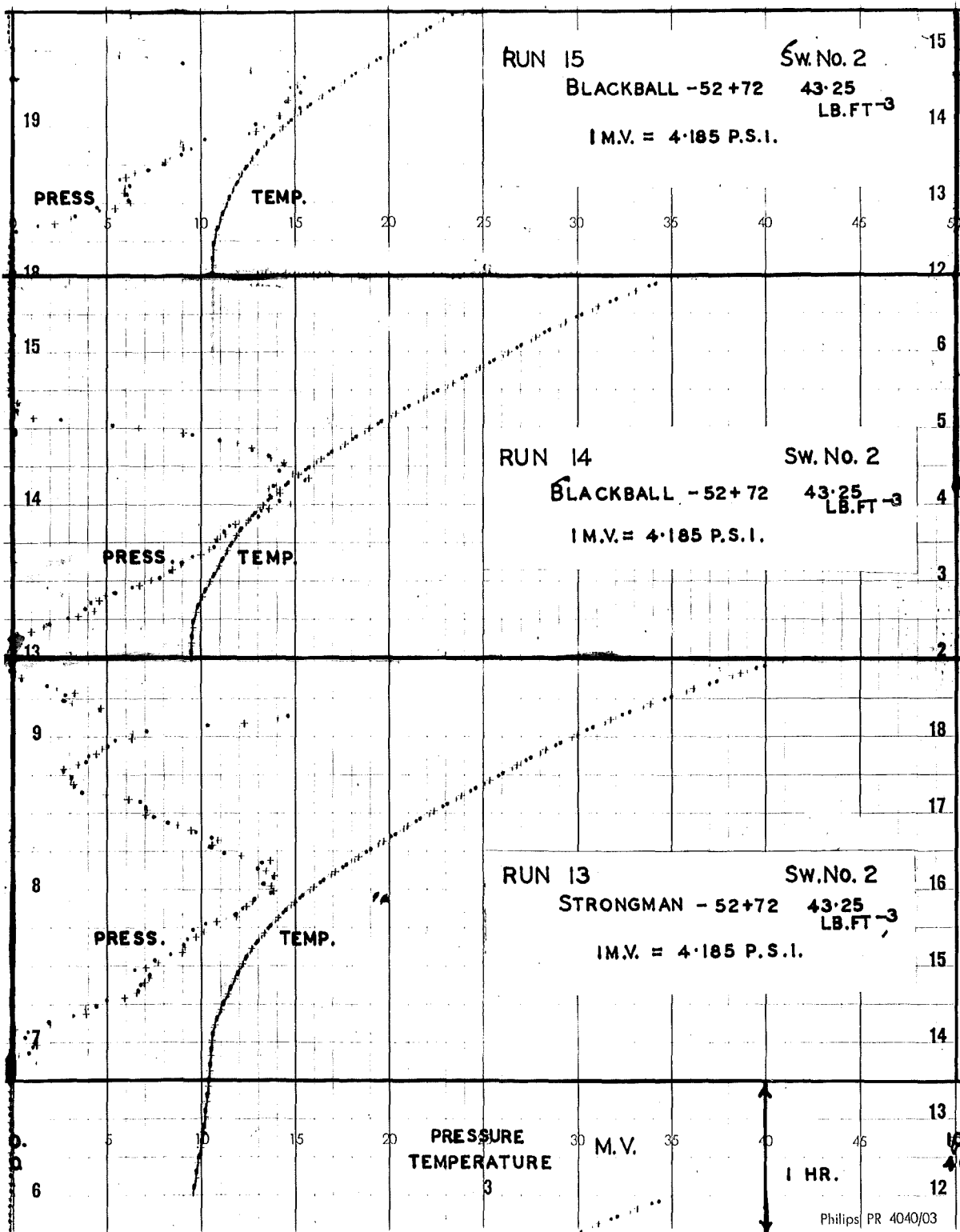
\*\* This sample was inadvertently preheated to 300°C. It was then cooled and the run proceeded as usual. The change in pressure characteristic could be due to preheating effects. No confirmatory runs were made. (op. Run 40 - subjected to long warm-up period.)

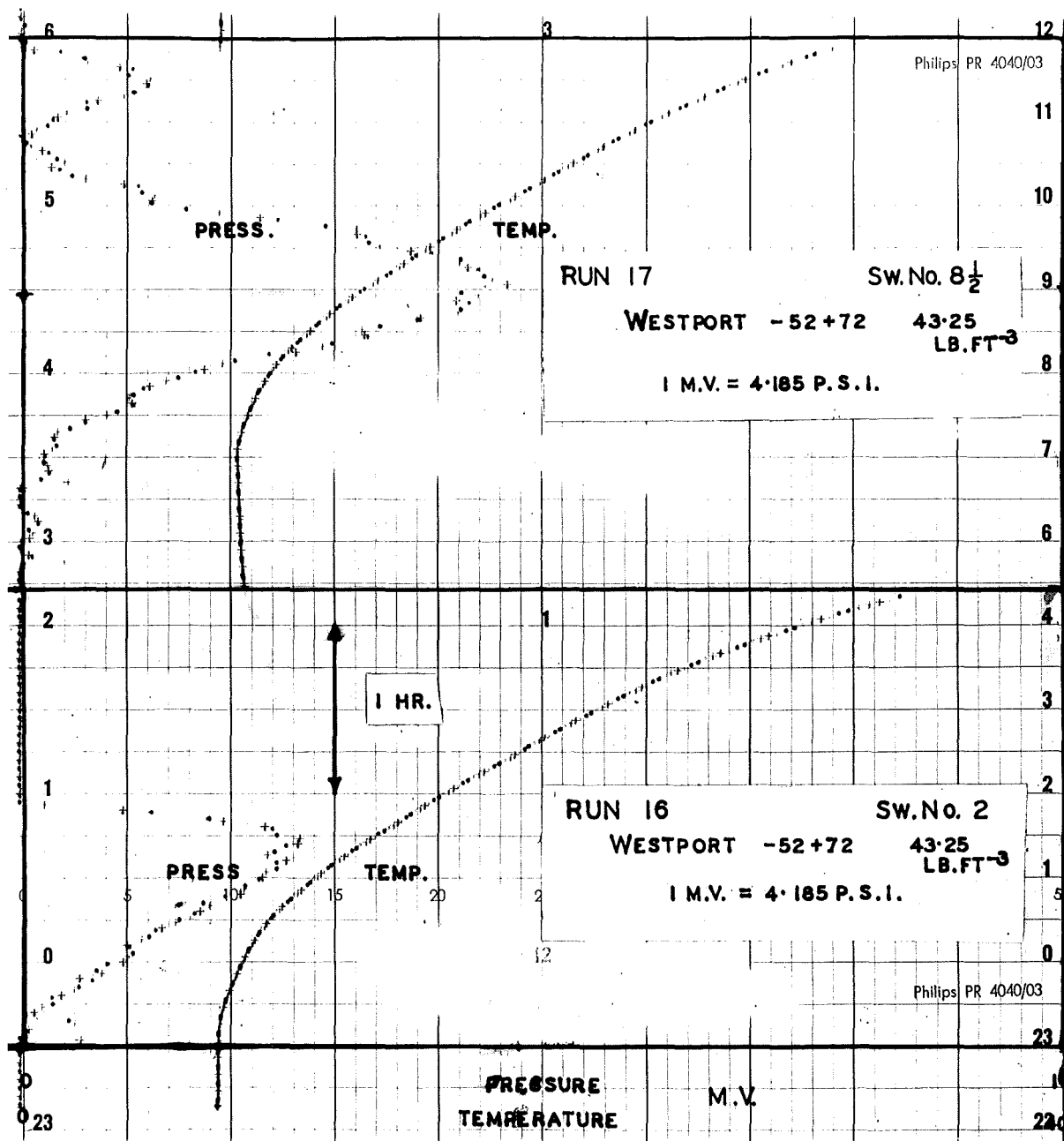




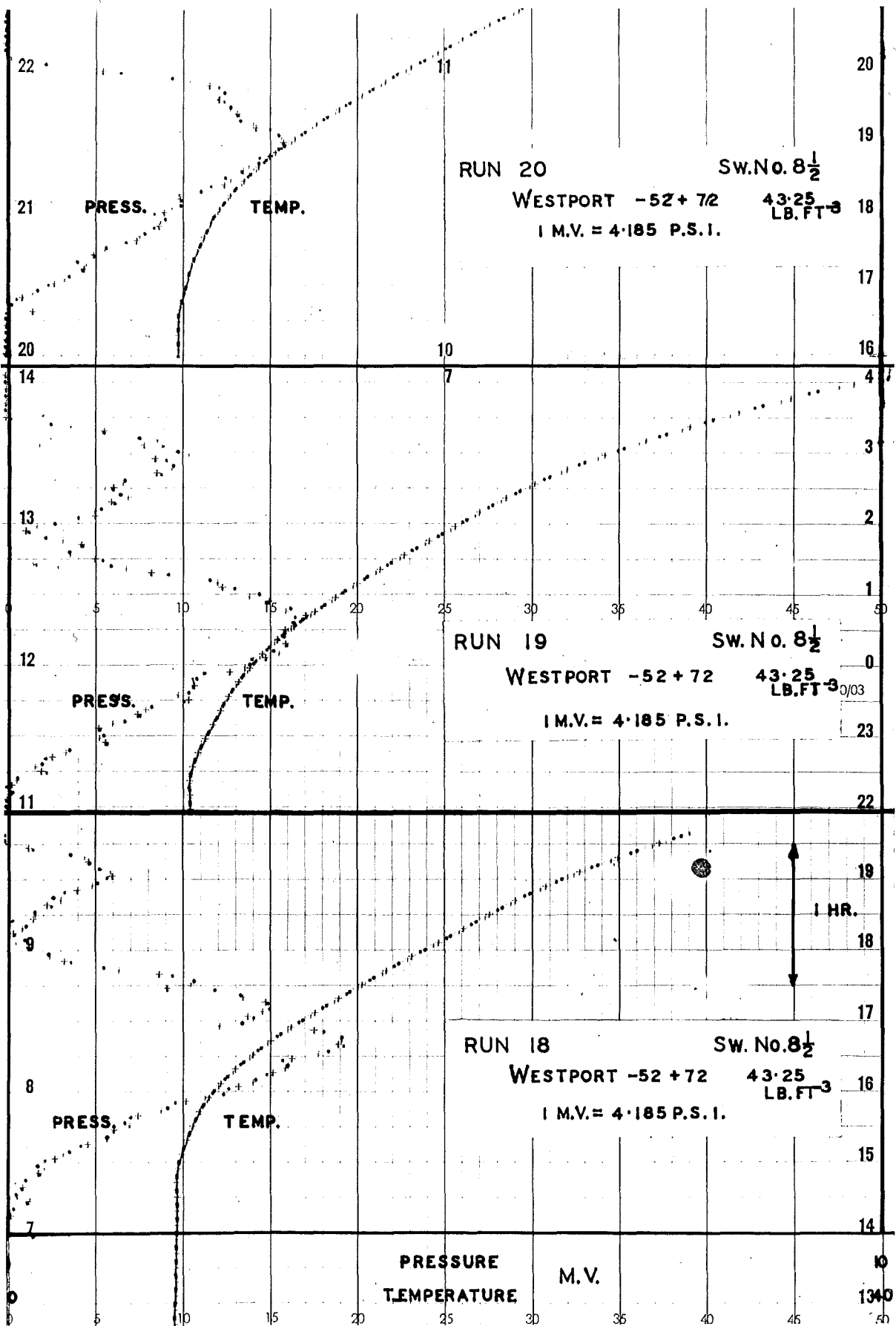


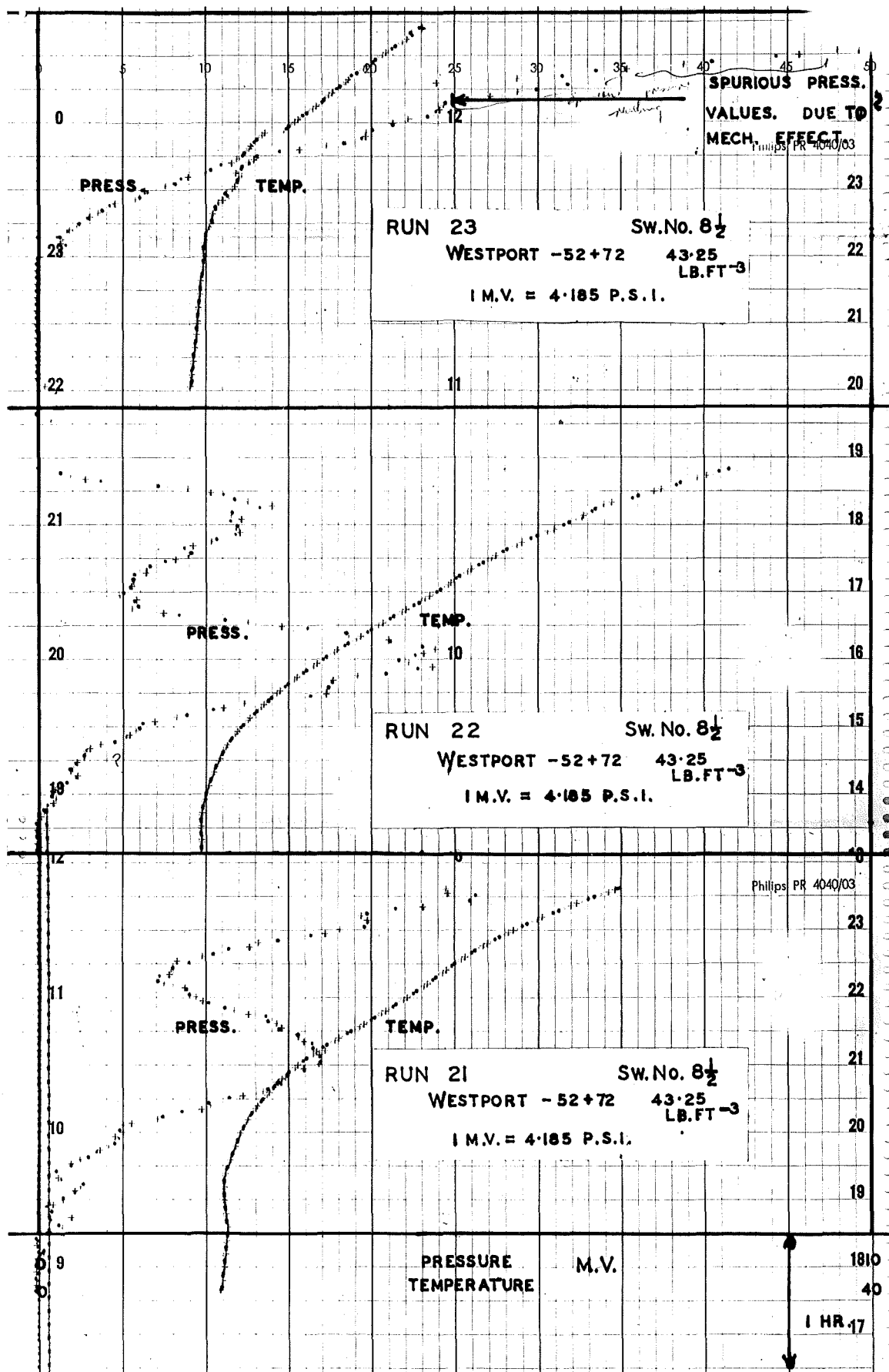


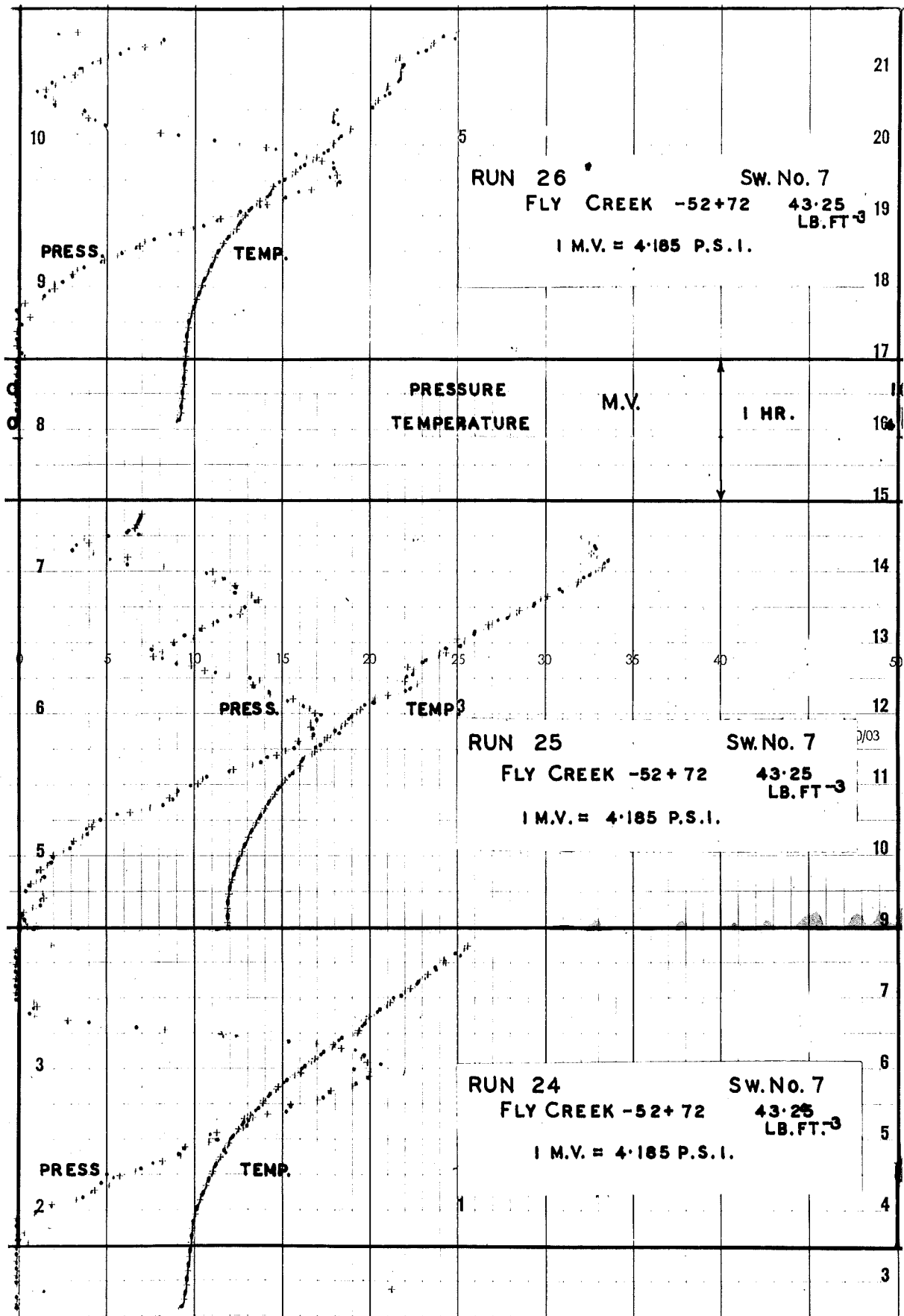


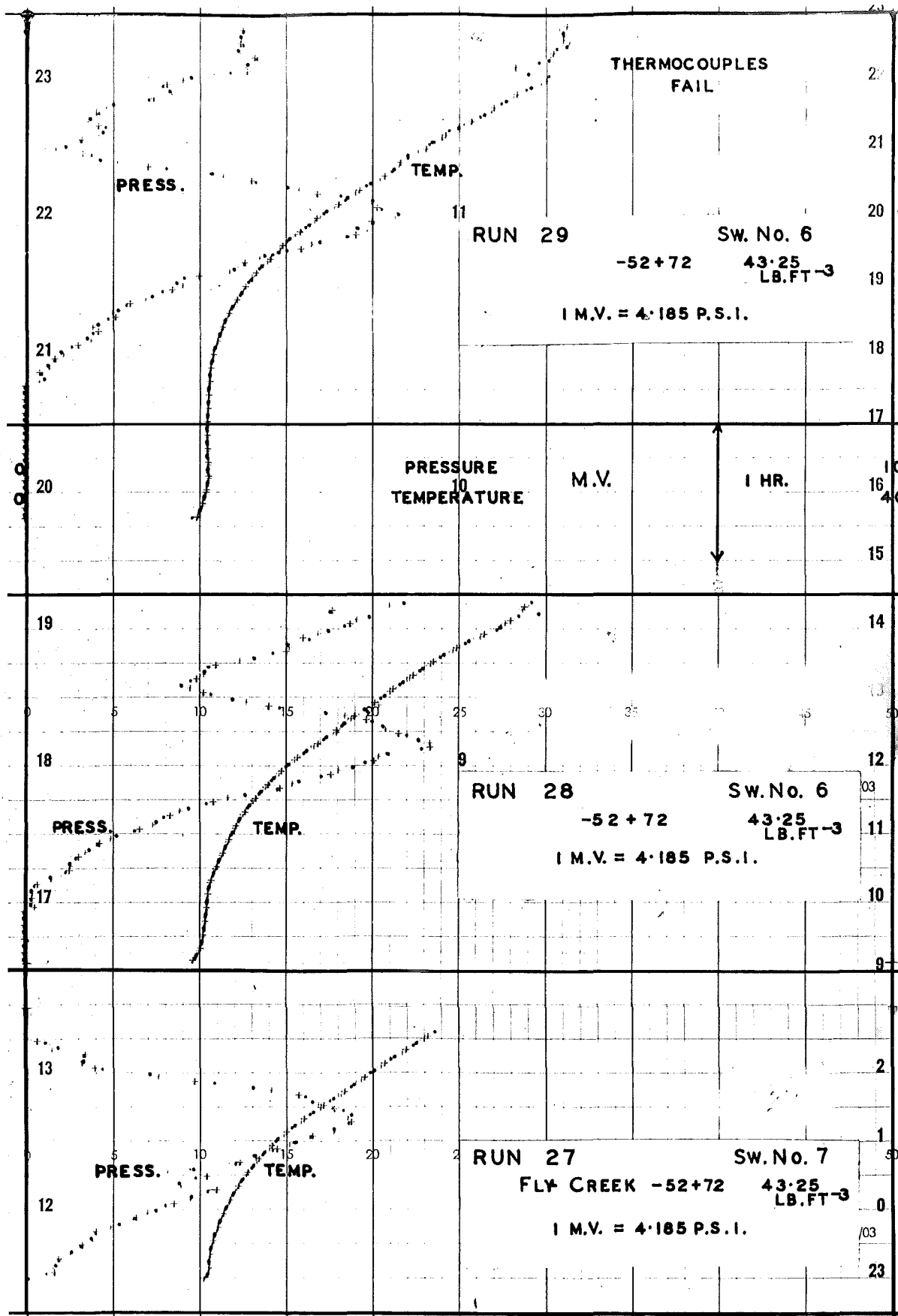


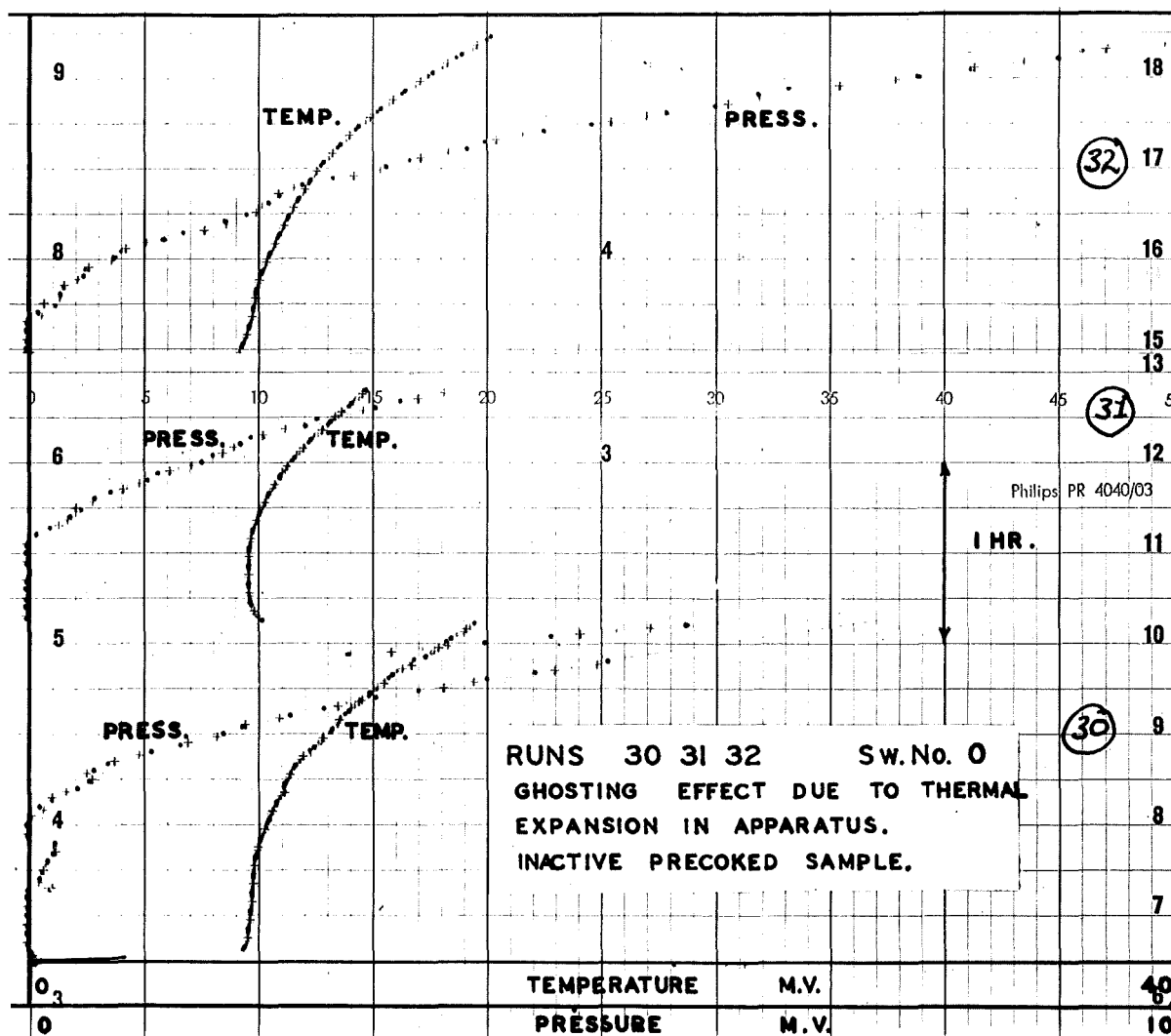


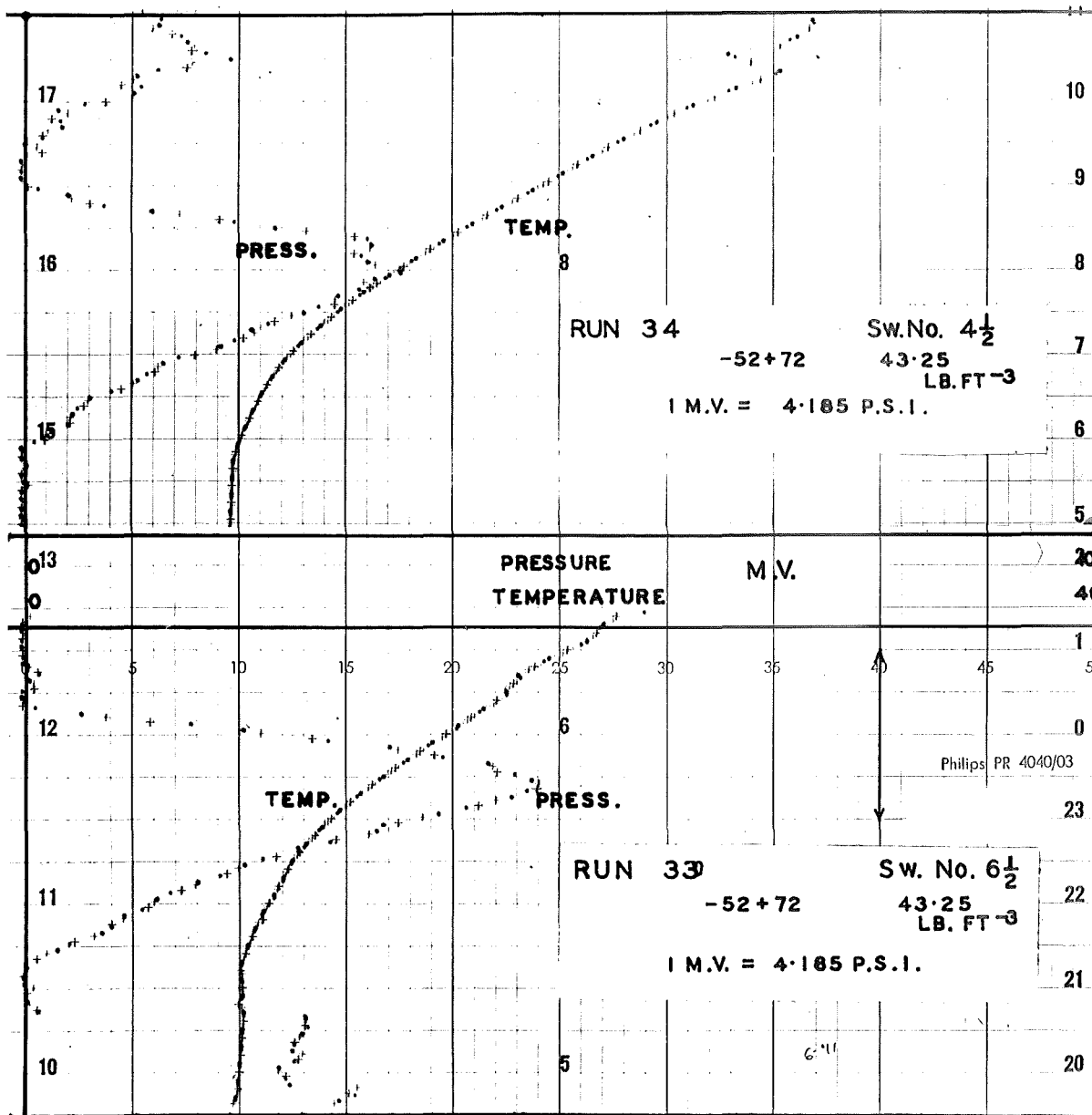


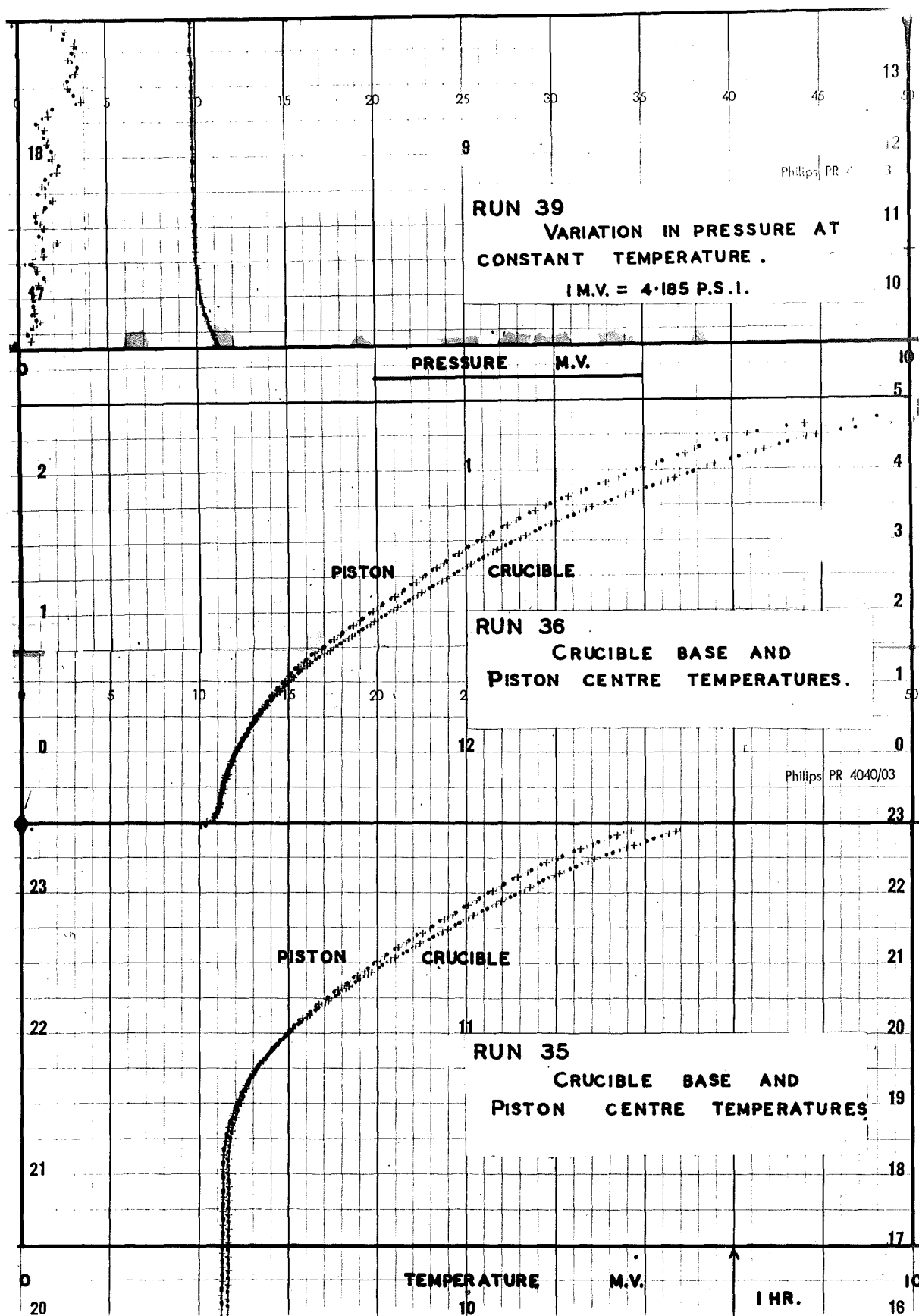


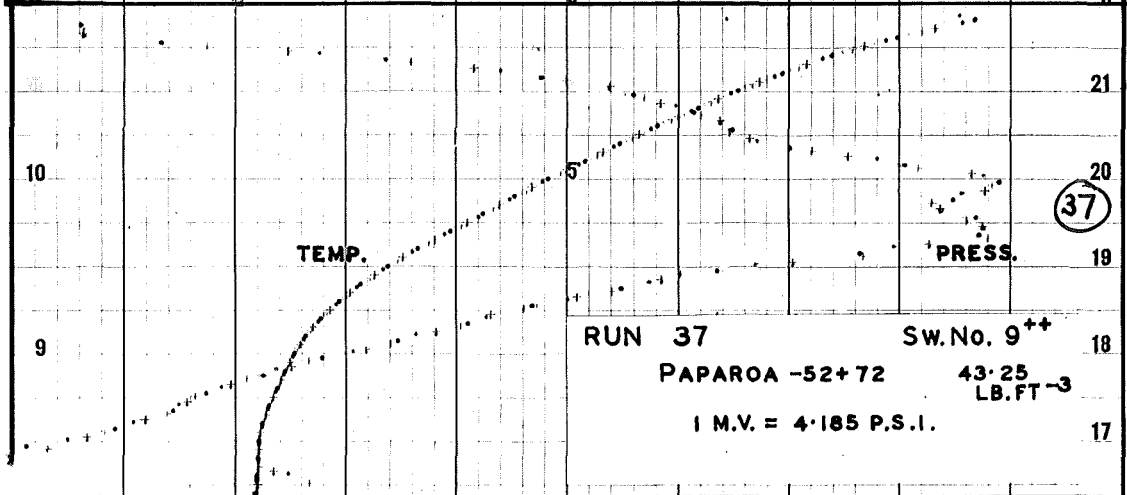
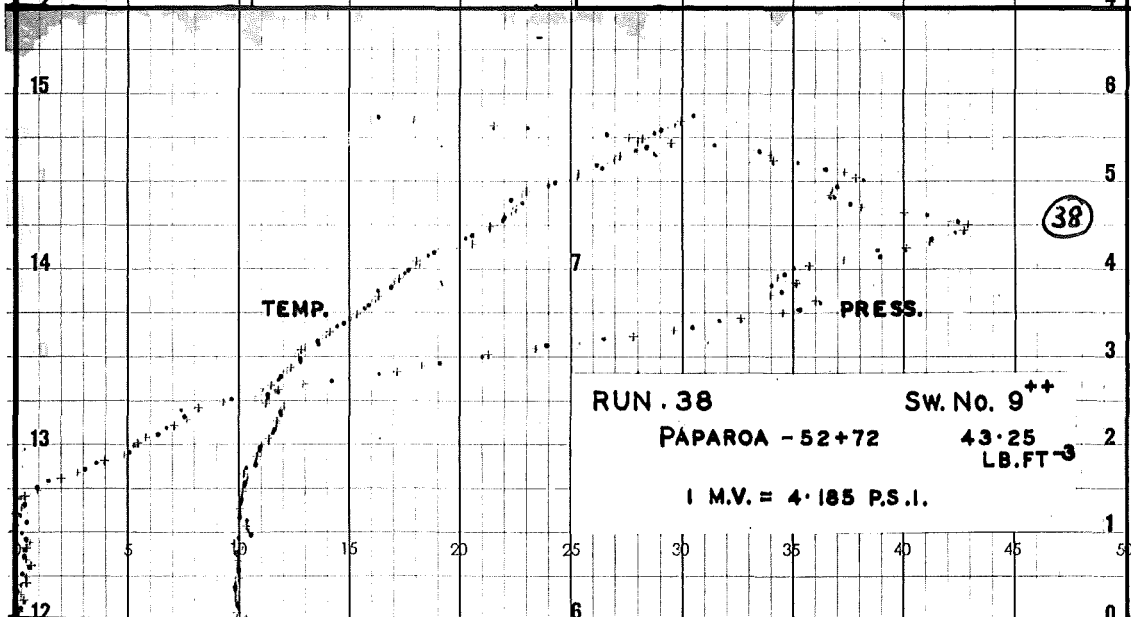
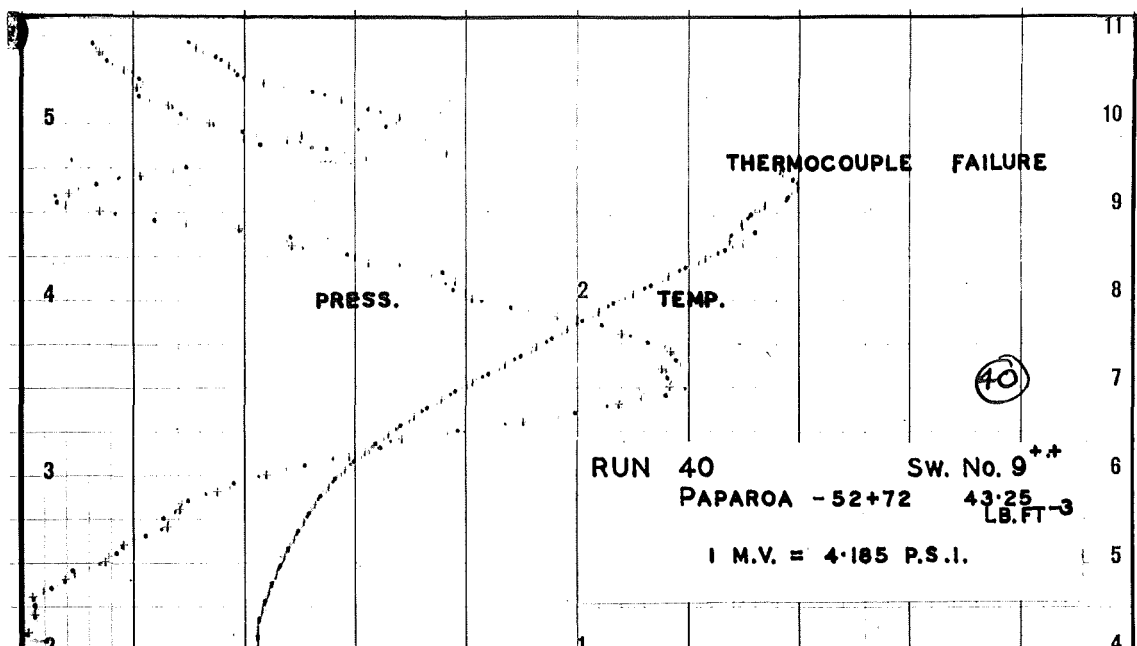














RESULTS

Tabulated results are shown in table 5. The recorded results from which the values tabulated have been taken are shown for the forty runs made in the photographic reproductions of the actual recorder chart RUN 1 - RUN 40. Original temperature is the constant temperature of thermal saturation of the furnace and crucible in the pre-run period. The columns characteristic temperature and characteristic pressure refer to the temperature and pressure values at the maximum point of the recorded pressure curve. These are characteristic of the apparatus and test procedure and are reproducible as long as standard apparatus is used and standard procedure followed. They are quantitative and relative values for swelling pressures of various coals as the apparatus effects are constant as long as standard procedure is used and standard conditions of temperature rise prevail.

The magnitude of apparatus effects was determined in runs 30-32 in which a button of coke from a previous run was put in the crucible in place of the coal sample. The columns headed corrected and actual pressure or temperature are the result of an endeavour to calculate absolute swelling pressures for the coals used and to find the temperatures at which these absolute pressures have a maximum value. The swelling numbers shown are those determined in crucible swelling tests carried out concurrently with the swelling pressure runs as described in the previous section.

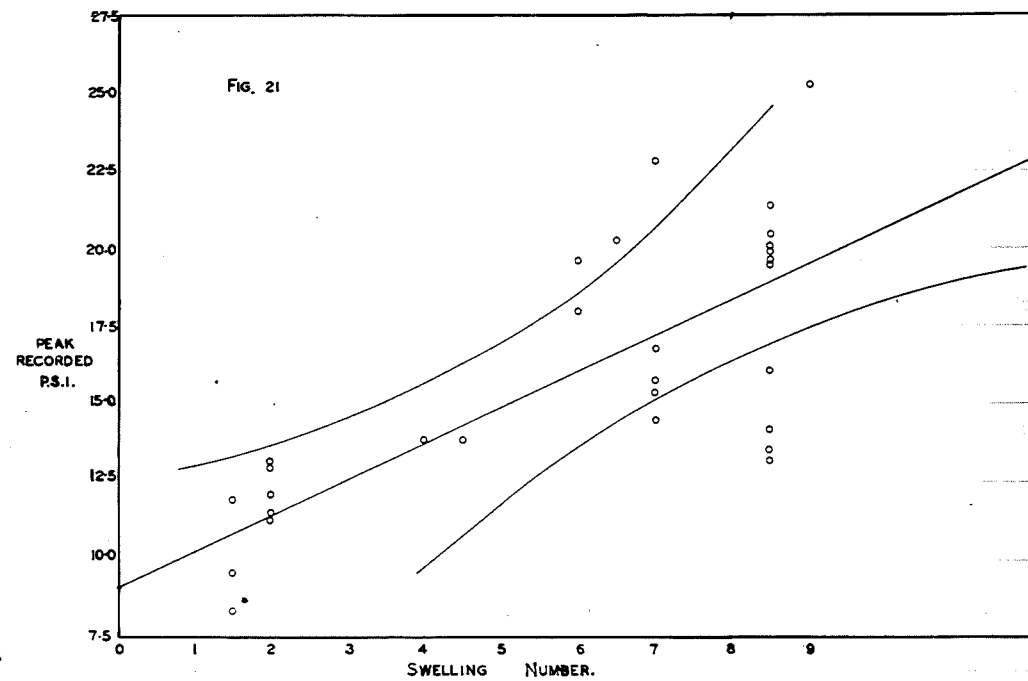
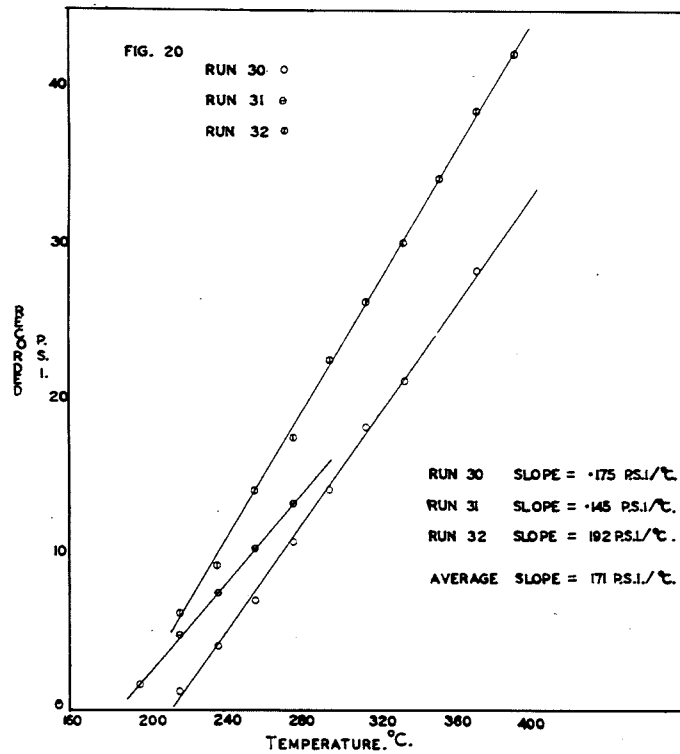
Samples Used

Some difficulty was experienced in obtaining coals of swelling numbers 3-6. It will be seen that some tests were made on mixed samples used because they were known to have swelling numbers in this range. The few

results available for coals of these swelling numbers leaves a weakness in the central range of the crucible swelling number scale, however, and makes correlation of the swelling pressure properties with the crucible swelling number difficult. Some runs have also been made on high swelling coal in order to compare the indicated equivalent swelling number so obtained with that predicted by Spooner's formula given previously. To assess the excess degree to which these coals swell in comparison with a coal of swelling number 9 was the initial purpose of the work done. These results also extend the rank range covered by the results obtained and are useful because of this in considering fundamental coal band properties.

Because the aim has been to establish correlations applicable to the New Zealand coal band as a whole, no attempt has been made to assess variations in petrography and their possible effect. It is thought that most New Zealand coals are petrographically similar (1); for a test of this nature to be of any real value any variations in petrography must have no biasing effect. Therefore coals from any source have been acceptable for subjection to the two tests. In fact all coals used have been from either Greymouth or Buller coalfields.

Similarly because the tests have been made simultaneously the state of ageing or weathering of the +7 mesh sample taken has not been of concern, it being assumed that this has the same effect on both properties measured. In table 5, where indication of increasing sample age is given this refers to the age of the -52 +72 and the -72 mesh samples prepared from the original +7 mesh sample. It seems from the two series of runs that such ageing tends to reduce the swelling pressure developed quite significantly. However, this reduction is not shown by the swelling number, an indication of the insensitivity of this test. Similarly preheating to elevated temperatures



or prolonged preheating at 200°C appears to alter the characteristic pressure development of a coal. These effects were not systematically investigated, the observations referred to being the result of failure to follow standard procedure in RUN 25 and RUN 40 respectively.

#### Influence of the Apparatus on Recorded Results

The apparatus effects present in the results as recorded were assessed by dummy runs with a pre-coked sample. Fig. 20 shows the pressure values recorded, taken from RUNS 30, 31, and 32, versus temperature rise from the original temperature. The average value of the slope is 0.171 psi/°C for the three runs. The effect is linear for all three runs which is to be expected as the effective pressure developed must be due to thermal expansion in the crucible support and piston rod. This effect emphasises the important effect of apparatus dimensions on the characteristic pressure values obtained and the shape of the recorded pressure curve. It is also the reason why any similar apparatus would have to be of identical construction if comparable results were to be obtained.

#### The Shape of the Recorded Pressure Curves

With the importance of this thermal expansion effect recognised the significance of the shape of the pressure curves obtained can be discussed. The initial positive pressure is found to be not wholly due to this thermal effect so the coal does develop a positive pressure. By calculation the magnitude of the pressure due to the coal sample can be obtained at any stage of the run up to the stage where the recorded pressure becomes zero again, i.e.

$$\text{Nett pressure in coal sample} = \text{Recorded pressure} - 0.171 (\text{Temperature} - \text{Original temperature}).$$

Performing such calculation gives the figures given in table 5 for corrected pressure - nett coal pressure at peak recorded pressure; and actual maximum pressure - the highest nett coal pressure developed.

The values in these columns show much more variation than the characteristic values as the calculation increases the percentage error greatly. However, the same general relationship in properties is evident in both the characteristic values and those derived from them in this way. The negative nett pressure values indicate that the samples have in effect shrunk and it is obvious from the shape of the curve that beyond the recorded maximum pressure this shrinkage effect is very sharp and of considerable magnitude.

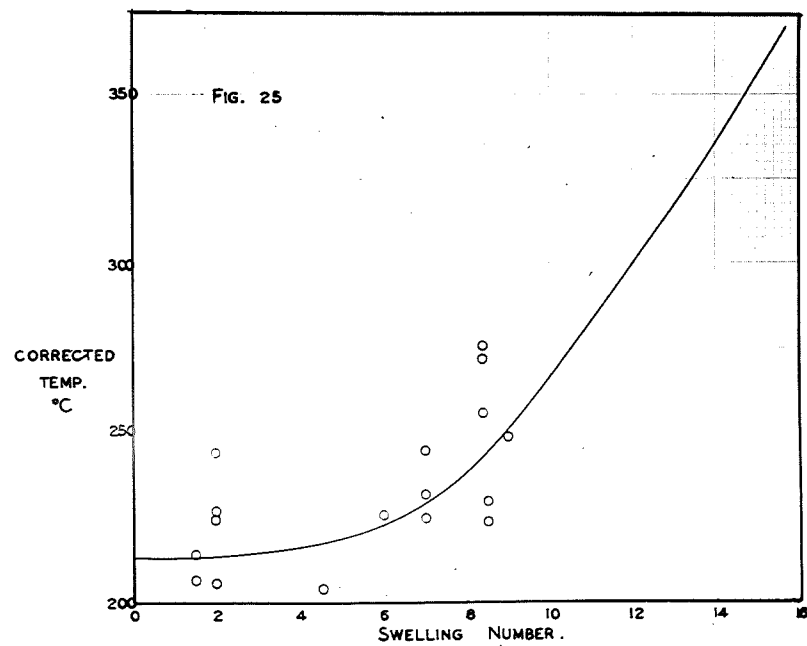
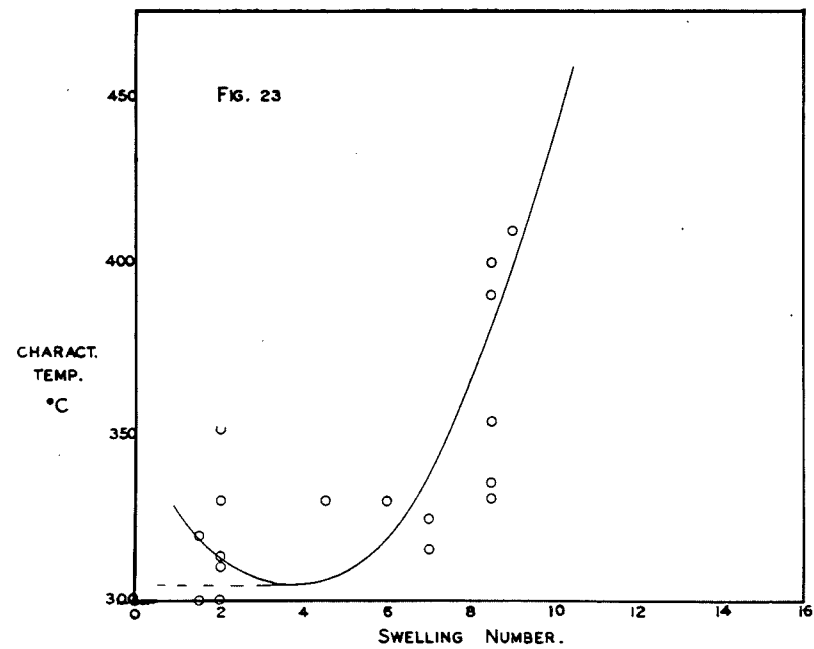
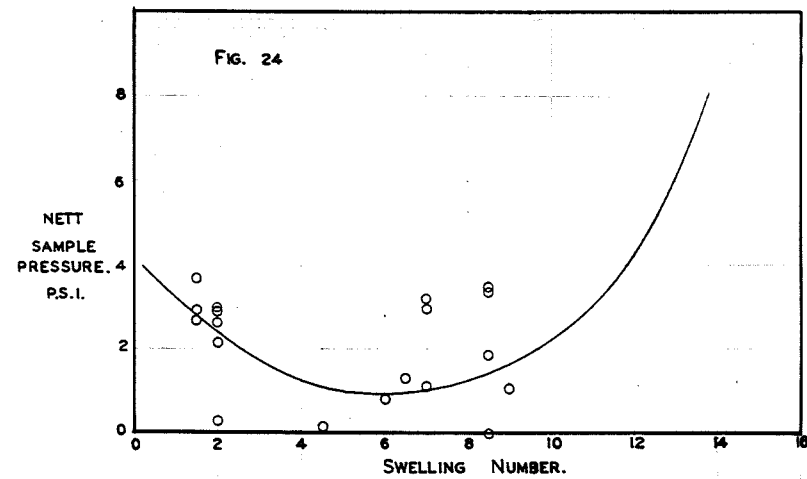
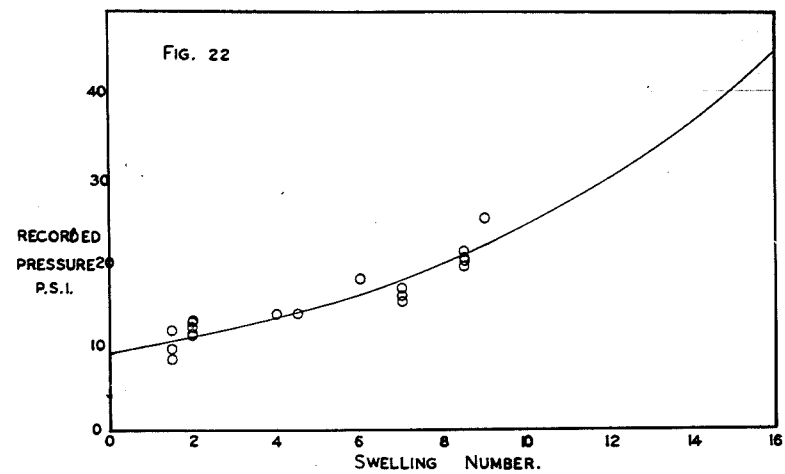
For most of the coals used the shrinkage effect reduces the recorded pressure to zero progressively from the maximum. However, for some coals an effect is observed, referred to in table 5 as the second peak, which indicates that at higher temperatures a change in rate of shrinkage occurs. Indeed for some coals the shrinkage would appear to stop for a time and then recommence, the recorded pressure going to zero at this stage. This second peak effect seems to be more evident in higher rank coals and is affected by ageing to a marked degree as shown in RUNS 17-20. The temperature at which this second peak occurs indicates that it could be due to the onset of a rigid coke formation in the crucible which prevents further shrinkage. The sudden recommencement of shrinkage may be due to a breakdown in this coke structure by the crushing action of the superimposed pressure. This would account for the granular non-rigidly coked condition of the sample at the end of a run, which seemed surprising in the case of strongly coking coals like Paparoa. In runs where no second peak occurs shrinkage prior to this coke formation is of sufficient magnitude that the

coke formation does not cause a positive recorded pressure to develop.

This second peak occurs at reproducible temperature values, but its magnitude varies considerably from run to run in some coals.

#### Comparison with Results of Other Workers

While no work of exactly the same nature as that undertaken has been found in the literature, results from similar apparatus used in investigating rather different problems can be compared with those presented here. Nedelmann (18), Baum and Heuser (19), and Koppens (20) have all worked with similar apparatus but in each case the sample has been larger and a temperature profile has been impressed on the sample. The measured quantity has been the pressure developed in the direction of travel of this temperature profile. Thus the pressure measured has not been significant of fundamental coal property changes during carbonisation. The results obtained by these workers show a maximum pressure developed but this is not related to sample temperature as no single temperature is representative of the sample state. Usually the nett pressure developed in the sample is plotted against time, the sample being introduced into a preheated furnace. Several workers have used percentage swelling under controlled heating as a measure of swelling properties including use of the Sheffield Laboratory Coking Test (21). Results from such tests in which once again large samples are used, are frequently given as plots of percentage expansion against temperature as the samples were heated at a linear rate of temperature increase of  $3-5^{\circ}\text{C}$  per minute. These results show a maximum swelling in the range  $400-450^{\circ}\text{C}$  which is a considerably higher temperature than has been observed for the maximum swelling pressure in the present work. The results given in the literature here cited are comparable with the



characteristic values given in table 5 as in every case they are characteristic of the particular apparatus and procedure and are not corrected for apparatus and procedure effects. Differences in these effects between the apparatus used could account for the differences in temperature at which maximum property magnitudes are observed. It is more likely, however, that the differences are due to the differences in observed property. It seems likely that swelling pressure and swelling are not directly related with respect to the temperatures at which they occur but that the pressure develops during softening and incipient plasticity while swelling occurs at the later stage of maximum fluidity and volatile evolution which Spooner (6) and Waters (12) have shown to occur together in the 400-500°C temperature range. It is interesting to note that Stott (13) found initial reaction under the oxidizing conditions used in his work to occur in the same temperature range as the maximum recorded pressures in this work. The suggestion that swelling pressure occurs during softening and incipient plasticity appears to be supported by this agreement in temperatures as the oxidation reactions can be considered to be favoured by the onset of fluidity. That swelling pressure is independent of volatile evolution is shown not only by the swelling pressures measured in an apparatus from which the volatiles had free escape but also by Brown (22) who showed that solidification occurred before maximum volatile evolution for medium and high rank coal.

Fig. 19 shows the rank dependence of the reactions which occurred in Stott's experiments. Figs 21-25 show the correlation of the pressure and temperature values of table 5 with crucible swelling number, which is effectively a rank parameter. The dependence of temperature on rank is in the same direction and of similar magnitude in the range considered.



Brown (loc. cit) also shows a temperature rank dependence for maximum and initial fluidity and maximum and initial volatile evolution of similar magnitude. The pressures also increase with rank. Sample ageing appears to cause an effective rank decrease which is shown by Suggate (1) to be the effect of weathering.

### Result Reproducibility

Values of characteristic pressure are reproducible under standard conditions to  $\pm 5\%$ . In fig. 21 the values for all runs are plotted, the linear regression line is shown and 90% confidence limits drawn in. All the values outside these confidence limits have been subject to non-standard conditions of testing, i.e. in sample age or in heating. All the other values listed in table 5 and shown in figs 22-24 are derived from this basic pressure measurement either by their relationship to it or by calculation and they are correspondingly less precise.

RUN 39 shows the error in pressure value recorded which can be expected from bridge variation, i.e.  $\pm 1.42$  psi. The percentage error introduced by this at low pressures is very great and combined with the apparatus effect could give a considerable resultant error in the actual pressure values obtained. From the slope values shown in fig. 20 it can be seen that an error of 10% could occur in the value of the apparatus effect estimated for a particular run. The temperatures at which these actual pressures occur are similarly less reproducible than the temperatures for peak recorded pressures.

The reproducibility of the temperature versus time curve has been commented on previously. It should be pointed out, however, that due to thermocouple failure after several runs it was necessary to replace the

thermocouple periodically. Such replacement had no noticeable effect on the shape of the curve or on the reproducibility of temperature values given in table 5. This indicates that no stray current effects influenced the temperatures recorded by the thermocouple and that the pressure effects observed did occur at what appear to be lower temperatures than may have been expected from the percentage swelling results previously referred to. This consideration is especially important in the case of nett swelling pressures in the coal. The results show that the maximum value for this pressure occurs at temperatures as low as  $210^{\circ}\text{C}$  which is much too low for volatile evolution or any of the changes accompanying it to be responsible for the swelling pressure. In the apparatus used the escape of gas is facilitated so this does not affect the pressure recorded.

A further demonstration of the accuracy of the temperatures recorded is given in RUNS 35 and 36. In the pre-run period the thermocouple in the piston shows a higher steady state temperature than that under the crucible. This indicates the effect on the indicated temperature from the crucible thermocouple of its hot junction position with respect to the crucible base. It was not actually embedded in the metal, only touching it. In comparison the piston thermocouple hot junction was brazed into the piston face, an integral part of the metal. At the steady state conditions existing in the pre-run period both temperatures could be expected to be the same. In fact the difference is  $6^{\circ}\text{C}$ . This difference can be considered to be a constant error in the temperature values recorded during runs. It is not significant, however, in discrediting conclusions that pressure effects occur at much lower temperatures than swelling effects do. Table 6 shows the values, taken from RUNS 35 and 36, for the temperatures at the two thermocouple positions as a run proceeds.

Table 6

RUN 36			RUN 35		
$T_1$	$T_2$	$T_1 - T_2$	$T_1$	$T_2$	$T_1 - T_2$
236.5°C	236.5°C	0°C	230.5°C	227°C	-3.5°C
259°C	261°C	2°C	242°C	238.5°C	-3.5°C
288°C	296°C	8°C	260°C	260°C	0°C
333°C	344.5°C	11.5°C	294°C	296°C	2°C
385°C	403°C	18°C	337°C	342°C	4.5°C
441°C	468.5°C	27.5°C	387°C	401°C	14°C
500°C	537.5°C	37.5°C	447.5°C	468°C	20.5°C
571°C	622°C	51°C	507.5°C	539.5°C	32°C
665.5°C	730°C	65.5°C	582.5°C	625°C	42.5°C
787°C	872°C	85°C			

These runs were made in the first place to endeavour to find the accuracy of the assumption that the sample was a unitemperate mass. It is evident that as the temperature increases this assumption becomes less valid. This is due partly to the establishment of transient heat conduction through the crucible sample assembly and the effects of changes in rate of heat loss from the sample area as the temperature increases. That the upper sample temperature lags behind the base temperature to a greater degree as the temperature increases is to be expected in view of Millard's (14) findings concerning the increasing degree of endothermicity of the carbonising reactions up to about 650°C. Differences between the two runs above are due to temperature differences at the commencement of the runs.

Conversion of the milli-voltage values recorded to temperatures was made using standard tables (7). The temperature corresponding to a given milli-voltage value was readily obtained to an accuracy of  $\pm 0.25^\circ\text{C}$ . The pressure recording system used was sensitive to  $\pm 0.25$  psi as discussed

under the section, Apparatus-Modification.

### The Effect of the Temperature Profile in the Sample

The increasing magnitude of the range of temperature in the sample has little effect on the usefulness or reproducibility of the characteristic pressure and temperature values obtained from the apparatus, as long as its effect on them is reproducible. It could, however, influence the absolute values obtained by attempts to eliminate such apparatus and procedure effects from the recorded results. In appendix 1 an analytical determination of the temperature profile detail is given. This shows that under the assumed conditions of the analysis the edge effects in the "flat" sample are small and that heating is effectively from the base of the crucible. Consequently the temperature profile effectively moves vertically from the crucible base through the sample to the piston. Under these conditions the swelling pressure measured is the maximum pressure in the sample as it has been shown by Koppers (loc. cit) and others that the swelling pressure is developed in the direction of travel of the temperature profile in the coal. The question to be decided is if this pressure is in effect the hydrostatic pressure within the plastic layer of coal during carbonisation or if it is merely a pressure characteristic of the coal being carbonised in the specified apparatus under the specified conditions. Brown (22) shows:

- (a) that the plastic layer in a carbonising coal mass is very narrow;
- (b) that the temperature profile through this plastic layer is very steep.

Nadziakiewicz while agreeing about the temperature gradient suggests a much thicker plastic layer occurs. As discussed previously, Nadziakiewicz and Callcott and Innes (23) used penetration plastometers which appear liable to give these greater thickness determinations because they do not differen-

tiate between the actively decomposing fluid zone and the softening and incipient decomposition of the coal on the colder face of the plastic layer. Considering these facts it seems reasonable to consider that the apparatus used here gives absolute pressure values which are the pressures actually developed in the plastic zone during carbonisation. The sample is 10 mm thick and the temperature gradient through the sample in the region of fluidity is not great enough for the sample to be considered as anything but a homogeneous small volume of coal being subjected to progressive carbonisation conditions.

#### Magnitude of Absolute Pressure Values

The absolute pressure values obtained range from zero to 8.00 psi with an apparent minimum - fig. 22 - at 4-5 on the Swelling Number Scale. This is a rather unexpected feature of the results obtained and could be due to the scant experimental values in this central region of the correlation of swelling pressure and swelling number. The values up to a swelling number of 8.5-9 do not exceed 3.50 psi. This is of interest in that the Christchurch Gas Company accepts this as the limiting coal swelling number for satisfactory operation in their intermittent vertical retorts. Lambert et al (24) in tests to destruction on commercial plant found that an actual pressure of 2.0 psi at the walls was limiting for safe operation. The values for absolute swelling obtained in the present work seem reasonable in the light of Lambert's work as bulk effects in commercial plant and the opportunity of limited plastic layer flow would almost certainly reduce the plastic layer pressure transmission to the walls.

In discussions with Christchurch Gas Company laboratory staff the author came to the opinion that the crucible swelling number apparatus

being used in their laboratory was not giving reproducible results as no standardisation of rate of heating was made. In tests on the same coals the author obtained buttons up to  $1\frac{1}{2}$ -2 swelling numbers greater than the Company's apparatus gave. In these tests the author's apparatus was standardised immediately prior to the test. This means that the Christchurch Gas Company could be carbonising coals of much greater swelling properties than coals of swelling number 9. The steepness of the swelling pressure increase in the  $>9$  region indicates, however, that  $9^{++}$  coals give pressures twice as great as those for 9 swelling number, so it is unlikely that  $9^{++}$  coals have ever been used as no serious damage has occurred to their plant. If the equivalent swelling numbers predicted by Spooner's formula, figs 6-8 in section II, are an accurate indication of relative swelling properties the  $1\frac{1}{2}$ -2 discrepancy would allow a  $9^{+}$  coal to be taken as a 9 coal however, which indicates that swelling pressures of 4.0-4.50 psi are tolerable, fig. 24. Hence if Lambert's 2.00 psi value at the wall is applicable to the Company's plant the reduction in transmitted pressure is considerably greater, up to 50%, if the absolute pressures given in table 5 are the pressures actually developed in the plastic layer.

#### Equivalent Swelling Numbers of $9^{+}$ $9^{++}$ and $9^{+++}$ Coals Predicted by Results

RUN 37 and RUN 38 are for Paparoa coal of swelling number  $9^{++}$ ; the values of pressure and temperature for these runs plotted on the correlating curves of figs 22-25 predict equivalent swelling numbers of  $14\frac{1}{2}$ ,  $10\frac{1}{2}$ ,  $13\frac{1}{2}$ , and 13 respectively. The average value is 13, a rather larger equivalent swelling number than is predicted by figs 6-8. The  $10\frac{1}{2}$  value makes this average as low as it is and it appears certain that the equivalent swelling numbers for  $9^{+}$   $9^{++}$  and  $9^{+++}$  are greater than 11,  $12\frac{1}{2}$ , and  $15\frac{1}{2}$ . In figs 6-8 the

plotted points for New Zealand coals do fall to the right of the curve and so these larger values may have been expected. The  $10\frac{1}{2}$  value predicted from fig. 23 is possibly the result of the considerable scatter of values at swelling number  $8\frac{1}{2}$  on this particular plot. These higher equivalent swelling numbers mean that the pressure reduction in commercial plant is even greater, or that the 2.0 psi value for dangerous wall pressure is not applicable to the plant considered here.

#### Correlation with BSS 1016 Swelling Number Scale

Figs 22-25 show respectively correlations of recorded peak pressure, temperature at which this pressure occurs, maximum nett pressure in the coal sample, and temperature at which this maximum pressure occurs with the Swelling Number Scale of BSS 1016 (2).

The values outside the 90% confidence limits of fig. 21 are for runs in which non-standard conditions were used. The corresponding values for these runs have been discarded in figs 22-25. Further analysis showed that the data of fig. 21 is more closely correlated by an exponential function, fig. 22, but that this makes no alteration to the usefulness of fig. 21 as a criterion for excluding from further consideration runs outside the 90% confidence limits. Correlations using higher powers of the independent variable - swelling number - give no improvement in correlation, table 6. Table 6 also shows the summarised correlation data from which the decision on the form of the correlations drawn in figs 23-25 was made. For all the correlated properties except recorded pressure third or fourth order functions give the best agreement with the plotted data.

Table 6

## SUMMARY OF CORRELATION DATA

Dependent Variable y	Equation of Correlation	Correlation Coefficient R	$\sqrt{\frac{(y-\bar{y})^2}{N-1}}$
Recorded Pressure			
fig. 21	$y = 7.75 + 1.64x$	0.91	2.28
	$y = 9.67 + 0.62x + 9.92x^2$		2.40
fig. 22	$y = 8.68 e^{0.10x}$	0.93	
Characteristic Temperature			
	$y = 285.55 + 9.61x$	0.73	26.79
fig. 23	$y = 351.55 - 25.46x + 3.42x^2$		16.66
Nett Sample Pressure			
	$y = 2.29 - 0.11x$	0.32	1.00
fig. 24	$y = 4.09 - 1.07x + 0.09x^2$		0.88
Corrected Temperature			
	$y = 199.61 + 5.41x$	0.85	10.00
fig. 25	$y = 215.44 - 2.99x + 0.82x^2$		9.38
	$y = 200 e^{0.02x}$	0.85	

Note:  $x$  = Swelling Number = Independent Variable.

However, the equations for powers of  $x$  greater than two are not satisfactory as correlations as they are influenced by data scatter having several inflexion points and turning points. Therefore the correlations shown are in every case of power less than or equal to two or exponential, according to which gives the best correlation.

In fig. 23 the equation in table 6 is shown as the continuous line. The turning point is due to the scatter of data at swelling number two and it is considered that the dotted line is the more likely form of the curve



if such scatter, due to experimental error, was not present, cf. fig. 25. The exponential functional relationship found for the recorded pressure is considered to be the result of the apparatus and procedure effects discussed previously. These effects are not present in the values of the other properties correlated.

It should be noted that the greatest contribution to the standard error terms comes from plotted data in the 4.5-8.0 swelling number range where there is a lack of data. The scatter and weighting effect of the many plotted points at swelling numbers of 2.0 and 8.5 are responsible for this.

## VI

CONCLUSIONS

The apparatus used appears to offer a means of quantitative assessment of the swelling properties of coal of much greater usefulness than the present Crucible Swelling Test. Some of the uncertainty evident in the forms of the pressure and temperature correlations with swelling number are attributable to the uncertainty associated with assessing the swelling numbers from the buttons obtained. This is especially true of successive runs with the same sample, when significant changes in swelling pressure occur but no observable change in swelling number.

Results obtained from the apparatus are comparable in reproducibility with those of other workers using similar testing devices and also with the results from any testing of such a complex substance as coal.

Deduced and calculated quantities are rather less reproducible, however, as they are subject to accumulated errors from several sources, the calculation often involving the differencing of quite large values to obtain a small resultant absolute figure. Despite these increasing errors correlating functions have been found which fit the data significantly, correlation coefficients of between 0.80 and 0.90 being obtained. The unprocessed results in terms of maximum recorded pressure correlate with swelling number exponentially with a correlation coefficient of 0.92. The other correlations are of second order in  $x$  - swelling number - and it is concluded that the swelling properties bear a second order <sup>quadratic</sup> reaction to swelling number, the exponential form of the other correlation being due to superimposed effects peculiar to the apparatus and test procedure. The shape of the volume increase versus swelling number curve is also a second order curve, fig. 6.

It has been shown that not only does the maximum pressure produced bear

this relationship to swelling number but also that the temperature at which swelling pressures reach a maximum increases with swelling number, i.e. with rank, as swelling number is a rank indicating property.

The temperatures at which maximum swelling occurs range from 200°C to 350°C. This dependence of temperature on rank is of significance in coal utilization problems such as carbonisation and briquetting especially if the development of swelling pressure is indicative of softening and incipient plasticity. The swelling pressures observed occur at temperatures much lower than was expected indicating that swelling pressure and swelling, though associated properties, do not occur together. Swelling occurs at a higher temperature than swelling pressure and is more dependent on volatile evolution. Swelling pressures appear at temperatures known to be only slightly above the softening temperatures and appear to be independent of volatile evolution through the plastic coal.

It has been shown that the period during which pressure is developed is followed by a contraction in the coal sample and then solid coke formation. This contraction is quite sudden and of considerable magnitude. This contraction of the charge in carbonisation plant could account in part for the reduction in pressure between the plastic layer and the wall. It is considered that the absolute pressures developed by the coal in the tests made is the pressure in the plastic layer during carbonisation. The values obtained for coals known to have been coked in commercial intermittent vertical retorts are twice as large as the accepted dangerous pressure at the walls of such plant. It is concluded that a considerable pressure reduction occurs due in part to the shrinkage mentioned and in part to permeation of the plastic zone into the micro-fissures and macro pores of the uncarbonised coal charge. A certain pressure reduction will also

occur because of the coals fluidity.

Correlations of pressures and temperatures measured with swelling number have been extrapolated beyond 9 on a linearly extended swelling number scale and from runs with a coal of 9<sup>++</sup> coke button the equivalent swelling number has been found to be 13. This value is thought to be lower than it might be because of one very low contributory estimate, attributed to data scatter and unequal weighting. The value 13-13 $\frac{1}{2}$  agrees with values predicted by formulae relating ultimate analysis and percentage swelling.

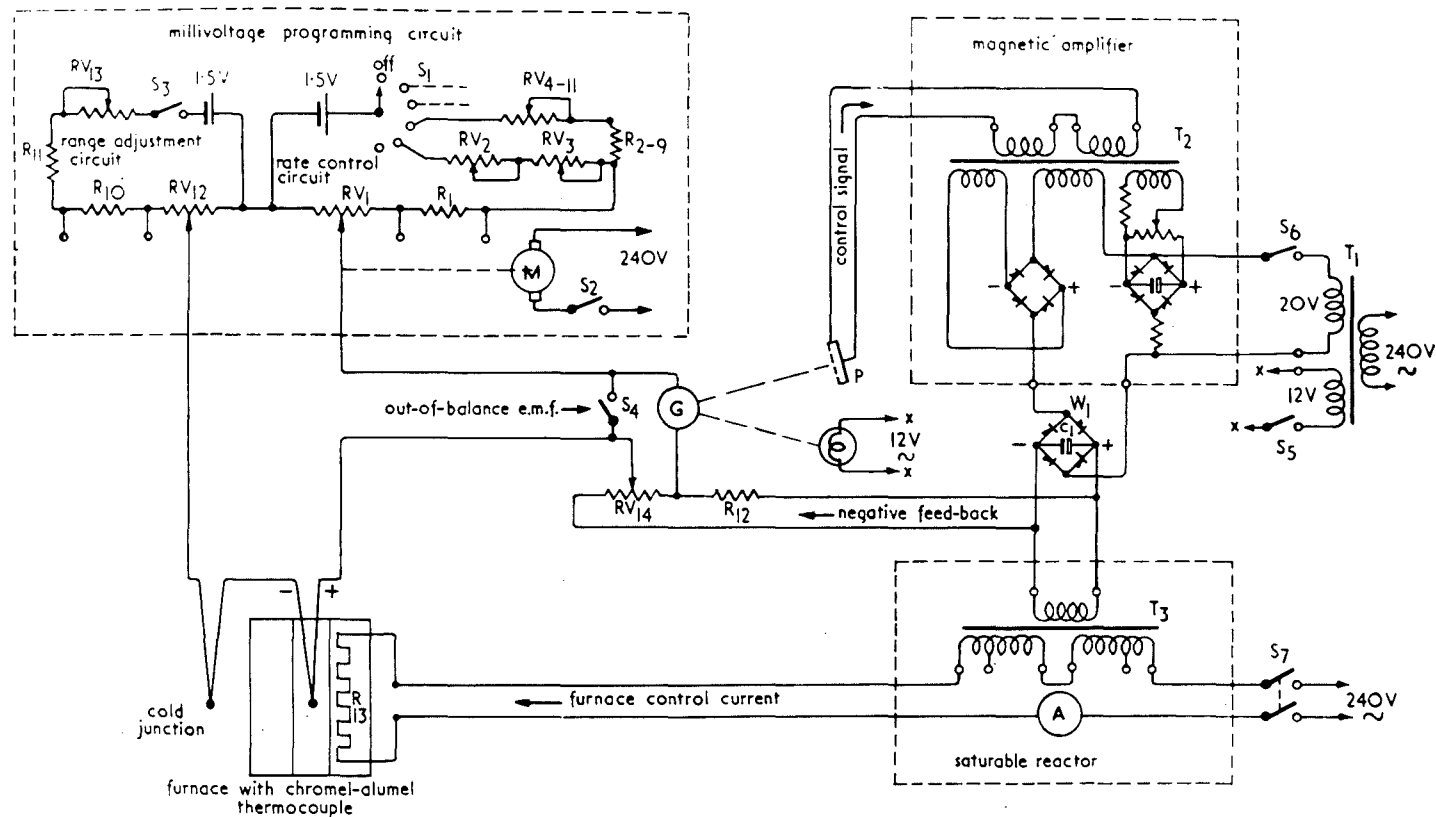


Fig.27. Circuit diagram of linear-rate temperature controller

$R_1$  2  $\Omega$   
 $R_2$  50  $\Omega$   
 $R_3$  15  $\Omega$   
 $R_4$  10  $\Omega$   
 $R_5$  50  $\Omega$   
 $R_6$  100  $\Omega$   
 $R_7$  50  $\Omega$   
 $R_8$  200  $\Omega$   
 $R_9$  300  $\Omega$   
 $R_{10}$  7.5  $\Omega$   
 $R_{11}$  600  $\Omega$   
 $R_{12}$  100 k $\Omega$   
 $R_{13}$  100  $\Omega$ , 600 W. furnace winding

$RV_1$  = 5  $\Omega$  slide-wire  
 $RV_2$  = 100  $\Omega$   
 $RV_3$  = 1500  $\Omega$   
 $RV_4$  = 50  $\Omega$   
 $RV_5$  = 50  $\Omega$   
 $RV_6$  = 100  $\Omega$   
 $RV_7$  = 100  $\Omega$   
 $RV_8$  = 200  $\Omega$   
 $RV_9$  = 250  $\Omega$   
 $RV_{10}$  = 500  $\Omega$   
 $RV_{11}$  = 500  $\Omega$   
 $RV_{12}$  = 20  $\Omega$ , 10-turn helical  
 $RV_{13}$  = 150  $\Omega$   
 $RV_{14}$  = 25  $\Omega$

$A$  = 0-5 A  
 $C_1$  = 50  $\mu$ F  
 $G$  = reflecting galvanometer, taut suspension; sensitivity, 1 mm  $\mu$ F at 1 m  
 $M$  = synchronous motor  
 $P$  = selenium photocell  
 $T_1$  = mains transformer for lamp and amplifier  
 $T_2$  = magnetic amplifier, type 560 (by Electro-Methods Ltd.)  
 $T_3$  = 1-kVA saturable reactor  
 $W_1$  = 1-kVA saturable reactor rectifier, type B45-1-1 (by Standard Telephones and Cables Ltd.)

## VII

THE COURSE OF FUTURE WORK

What has been done with the apparatus thus far opens up the possibilities of both immediate application as a swelling property measuring device and also use in conjunction with other techniques in elucidating the course of softening, plasticity, volatile evolution and coke formation in a mass of coal subjected to carbonisation.

The apparatus could be used without further modification for the first application, peak recorded pressure being used as the criterion of swelling properties.

To make it of maximum value in the more fundamental application, the elimination of the thermal expansion effect on recorded pressure values from the apparatus would be desirable. Recorded pressures would then be the pressure due to the coal alone and the uncertainty involved in calculation would be eliminated.

Identification of the changes occurring at a particular point in the pressure curve could be attempted using solvent extraction and plastometer techniques. Such techniques may similarly elucidate the rank dependence of swelling properties found in this work.

The apparatus could also be used in conjunction with carbonising tests to determine the effect of bulk density, imposed pressure and rate of heating on the quality of coke obtained. The effect of rate of heating would require a major modification in the form of a variable constant rate temperature controller like that used by the C.S.I.R.O., fig. 27.

## VIII

SYMBOLS USED

A	-	constant
	-	cross-sectional area, in <sup>2</sup>
B	-	constant
E	-	Young's Modulus, lb in <sup>-2</sup>
H	-	% composition hydrogen
k	-	gauge constant
	-	coefficient of thermal conductivity
N	-	number of observations
O	-	% composition oxygen
P	-	pressure, psi
R	-	resistance, ohms
	-	coreclation coefficient
T	-	temperature, °C
t	-	time, hrs
x	-	independent variable swelling number
y	-	dependent variable
$\bar{y}$	-	mean value of y
$\alpha_t$	-	temperature coefficient of resistance, ohms/ohm/°C
$\epsilon$	-	sensitivity
$\eta$	-	fraction of total time, secs/sec
$\Delta P$	-	force, lbs

## IX

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## APPENDIX 1

### Temperature Distribution in the Sample

The following is an attempt to determine the shape of the temperature profile in the sample, referred to earlier. The relaxation solution has been done for the sample when the crucible base temperature is  $400^{\circ}\text{C}$  and the temperature difference across the sample is  $15^{\circ}\text{C}$ . This is also in the temperature range at which the swelling and pressure properties are of interest and it is known that fluidity is approaching a maximum. However, the temperature profile will be similar at different overall temperature differences.

A network of radial lines and eight concentric circles has been considered. Fig. 26 shows the cross-section through the sample with relaxation temperatures in one half of the figure and isotherms drawn in the other. It has been assumed that:

- (1) the temperature at a particular value of  $n$  and  $h$  is the same at any such cross-section - giving only four temperatures in the left-hand side of the relaxing equation for  $T_0$ , the temperature at an interstice of the network.
- (2) the metal surfaces in contact with the sample are isothermal surfaces, the sides and base being at the crucible base temperature, the upper surface at the piston centre temperature. This is done in view of the high  $k$  values for mild steel and cast iron and the fact that an air gap exists between crucible and piston at A - in fig. 26.
- (3) the coefficient of thermal conductivity  $k$  has been assumed constant and, as implied in (2), significantly smaller than  $k$  for cast iron or steel.

For the network  $r_m = nx \pm \frac{x}{2}$  for the cross-sectional area  $A_r (= r_m \theta)$  between grid interstices in the radial direction of heat transfer and  $L_r = x$

in this direction. For vertical heat transfer  $A_v = nx^2\theta$ ,  $L_v = x$ .  
The characteristic grid length  $x$  is taken as  $\frac{2}{32}$  in.

The relaxing equation is then given by

$$\frac{k(T_1 - T_0)(nx - \frac{x}{2})\theta}{x} + \frac{k(T_2 - T_0)(nx + \frac{x}{2})\theta}{x} + \frac{k(T_3 - T_0)(nx^2\theta)}{x} + \frac{k(T_4 - T_0)nx^2\theta}{x} = 0$$

$$\text{i.e. } (T_1 - T_0)(n - \frac{1}{2}) + (T_2 - T_0)(n + \frac{1}{2}) + nx(T_3 - T_0) + nx(T_4 - T_0) = 0$$

$$\text{i.e. } nx(T_3 + T_4) + n(T_1 + T_2) + \frac{T_2 - T_1}{2} = T_0(2n + 2nx)$$

APPENDIX 2A Formula Relating Ultimate Analysis with Swelling Properties for  
New Zealand Coals

Efforts to find a formula similar to that of Spooner, p.4, but correlating ultimate analysis of New Zealand coals to percentage swelling, showed that no satisfactory formula existed. It is thought that the problem lies in the adoption of Spooner's percentage swelling figures at given crucible swelling numbers as the basis for derivation. As discussed previously the swelling test was devised for British coals and the repression of free swelling at and above swelling numbers of  $6\frac{1}{2}$  seems to be more important for New Zealand coals. For a similar formula to be obtained a basis of percentage swelling arrived at from considering New Zealand coals swelling freely, i.e. not coke button volumes, would seem necessary.